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DETERMINATION OF SIZE DISTRIBUTION (SDF)
FUNCTION BY ELECTROOPTICAL METHOD

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DETERMINATION OF SIZE DISTRIBUTION FUNCTION (SDF)

BY ELECTROOPTICAL METHOD

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In this work a method of determination of size distribution of colloidal particles has been developed. For comparison with other methods, a review work is presented in the synopsis part and the pertinent experimental results have also been presented.

ABSTRACT

When particles are in polare medium the molecules of the liquid will absorb on the surfaces of the particles. The resulting permanent electric dipole moment is used as the specification of the size of particles. Using the behavior of anisotropical particles in a rotating electric field, the SDF is determined for polydisperse system of particles.

CHAPTER 1

SYNOPSIS

1.1 SCATTERING OF ELECTROMAGNETIC WAVE BY SPHERICAL PARTICLES

The vector wave equations of electromagnetic wave propagating in anisotropic media are:

$$\text{Curl } \vec{E} + \frac{1}{C} \frac{\partial B}{\partial t} = 0 \dots\dots\dots (1-111)$$

$$\text{Curl } \vec{H} = \frac{1}{C} \frac{\partial D}{\partial t} + \frac{4}{C} \pi \vec{J} \dots\dots\dots (1-112)$$

$$\text{Div } \vec{B} = 0 \dots\dots\dots (1-113)$$

$$\text{Div } \vec{D} = 4\pi\rho \dots\dots\dots (1-114)$$

Using constitutive law and equations above the vector wave equations which could be derived are;

$$\Delta \vec{E} + \sigma \frac{\partial \vec{E}}{\partial t} + \frac{\partial^2 \vec{E}}{C^2 \partial t^2} = 0 \dots\dots\dots (1-115)$$

$$\Delta \vec{H} + \frac{\partial \vec{H}}{\partial t} + \frac{1}{C} \frac{\partial^2 \vec{H}}{\partial t^2} = 0 \dots\dots\dots (1-116)$$

where σ and c are the conductivity of the medium and the speed of light respectively. For the field quantities written in the form;

$$\vec{A} = (\vec{\alpha} + i\vec{\beta}) \text{ EXP } (i\omega t) \dots\dots\dots (1-117)$$

the Maxwell's equations reduce to the form;

$$\text{Curl } \vec{H} = ikm^2 \vec{E} \dots\dots\dots (1-112a)$$

$$\text{Curl } \vec{E} = -ik \vec{H} \dots\dots\dots (1-111b)$$

Where, $m^2 = \omega - i \frac{4\pi\sigma}{\omega}$ (1-118)

and k is the propagation constant in Vacuum. The solution of equation (1-118) is given by;

$$m_n = \sqrt{m^2} \left[\cos\left(\frac{2\pi n + \theta}{2}\right) + i \sin\left(\frac{2\pi n + \theta}{2}\right) \right], n = 0, 1$$

$$\text{and } \tan \theta = \frac{4\pi\sigma}{\varepsilon\omega}$$

For homogeneous media equations 1-111b and 1-112a should have solutions whose components satisfy the wave equation;

$$\Delta \psi = -k^2 m^2 \psi \text{ (1-119)}$$

For a plane wave travelling along the Z-direction ψ has the form, $\psi = \exp(i(-kmz + \omega t))$, Where km is the propagation constant in the medium with refractive index m . The wave will be damped if m has a negative imaginary part and is undamped if m is real.

In the latter case $\lambda_m = \frac{\lambda_0}{m}$. For propagation through two different media of refractive index m_1 and m_2 the boundary conditions used to solve the differential equations are; $\vec{n} \times (\vec{H}_2 - \vec{H}_1) = 0$ and $\vec{n} \times (\vec{E}_2 - \vec{E}_1) = 0$, which hold for the tangential components. For normal components the boundary conditions are;

$$\vec{n} \cdot (m_2^2 \vec{E}_2 - m_1^2 \vec{E}_1) = 0 \text{ and}$$

$$\vec{n} \cdot (\vec{H}_2 - \vec{H}_1) = 0, \text{ where } \vec{n} \text{ is the unit normal vector from the}$$

direction of propagation to the interface (6). Based on these arguments the solutions of the wave equation could be obtained.

It is assumed, as in most usual cases, that the scalar wave equations are separable into spherical coordinates and because of the

additivity of the scattered intensity, assumes solutions of the type;

$$\psi_{ln} = \begin{bmatrix} \cos(l\phi) \\ \sin(l\phi) \end{bmatrix} P_n^l(\cos\phi) Z_n(mkr) \dots (1-120)$$

where ϕ is the azimuthal angle.

In equation (1-120) n and l are integers and $n > l > 0$. $P_n^l(\cos\theta)$ is associated legendre polynomial and $Z_n(\rho) = \sqrt{\frac{\pi}{2}} Z_{n+\frac{1}{2}}(\rho)$ is the spherical bessel function. By virtue of equations (1-115) and (1-116), the field vectors $\vec{H}$ and $\vec{E}$ must be solutions of $\Delta \vec{A} + k^2 \vec{A} = 0$. If further ψ satisfies the scalar wave equation then; $\vec{M}_\psi = \text{Curl}(\vec{r}\psi)$ and $mk\vec{N}_\psi = \text{Curl} \vec{M}_\psi$, should also satisfy the vector wave equations. In addition if U and V are two independent solutions of the scalar wave equation and $\vec{M}_U$, $\vec{N}_U$, $\vec{M}_V$ and $\vec{N}_V$ are the derived vector fields, the Maxwell's equations will be satisfied by;

$$\vec{E} = \vec{M}_V + i\vec{N}_U \dots (1-121)$$

$$\vec{H} = m(-\vec{M}_U + i\vec{N}_V)$$

Equation (1-121) shows that the components of $\vec{E}$ and $\vec{H}$, should be written in terms of the scalar solutions U and V . In the case of a homogeneous sphere an assumption is made such that $m_2 = 1$ and $m_1 \neq 1$ and also the field quantities be expressed as;

$$\vec{E} = \vec{A}_x \exp(-ikz + i\omega t) \text{ and } H = A_y \exp(-ikz + i\omega t)$$

U and V can be obtained as;

$$U = \exp(i\omega t) \cos\phi \sum_{n=1}^{\infty} (-1)^n \frac{2n+1}{n(n+1)} P_n^1(\cos\theta) Z_n(kr)$$

$$\text{and } V = \exp(i\omega t) \sin\phi \sum_{n=1}^{\infty} (-1)^n \frac{2n+1}{n(n+1)} P_n^1(\cos\theta) Z_n(kr)$$

The scattered wave, hence obtained outside the sphere is given by;

$$U = \exp(i\omega t) \cos\phi \sum_{n=1}^{\infty} -a_n (i)^n \frac{2n+1}{n(n+1)} P_n^1(\cos\theta) h_n^{(2)}(kr)$$

$$V = \exp(i\omega t) \sin\phi \sum_{n=1}^{\infty} -b_n (i)^n \frac{2n+1}{n(n+1)} P_n^1(\cos\theta) h_n^{(2)}(kr) \dots (1-122)$$

Equation (1-122) is the modified form of equation (1-120) and

$$h_n^{(2)}(kr) = \frac{i^{n+1} \exp(-ikr)}{kr}.$$

Using the boundary conditions after representing

$$\psi_n(z) = z Z_n(z) = \left(\frac{\pi z}{2}\right)^{\frac{1}{2}} Z_{n+\frac{1}{2}}(z) = S_n(z)$$

$$X_n(z) = -z N_n(z) = -\left(\frac{\pi z}{2}\right)^{\frac{1}{2}} N_{n+\frac{1}{2}}(z) = C_n(z)$$

$$y_n(z) = z h_n^{(2)}(z) = \left(\frac{\pi z}{2}\right)^{\frac{1}{2}} H_{n+1}^{(2)}(z)$$

and taking the argument $X = ka$, $y = mka$, where a is the radius of the sphere; the coefficients a_n and b_n then obtained are,

$$a_n = \frac{\psi_n(y) \psi_n'(x) - m \psi_n(x) \psi_n'(y)}{\psi_n'(y) y(x) - m \psi_n(x) y'(y)}$$

and

$$b_n = \frac{m \psi_n'(y) \psi_n(x) - \psi_n(y) \psi_n'(x)}{m \psi_n'(y) y_n(x) - \psi_n(y) y_n(x)}$$

the scattered field at a large distance is approximated by the asymptotic behaviour of equation (1-122) to be;

$$U = \frac{-i \cos\phi}{kr} \exp(ikr + i\omega t) \sum_{n=1}^{\infty} a_n \frac{2n+1}{n(n+1)} P_n^1(\cos\theta)$$

$$V = \frac{-i \sin\phi}{kr} \exp(-ikr + i\omega t) \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} P_n^1(\cos\theta) b_n$$

Therefore, the tangential field components E_θ and H_θ can be derived by designating;

$$\pi_n(\cos\theta) = \frac{1}{\sin\theta} P_n^1(\cos\theta)$$

and

$$\tau_n(\cos\theta) = \frac{dP_n^1(\cos\theta)}{d\theta}$$

$$\text{as; } E_\theta = H_\theta = \frac{-i}{kr} \exp(-ikr+i\omega t) \cos\phi S_1(\theta)$$

$$-E_\theta = H_\theta = \frac{-i}{kr} \exp(-ikr+i\omega t) \sin\phi S_2(\theta)$$

where

$$S_1(\theta) = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} [a_n \pi_n(\cos\theta) + b_n \tau_n(\cos\theta)]$$

$$S_2(\theta) = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} [b_n \pi_n(\cos\theta) + a_n \tau_n(\cos\theta)]$$

As the radial component decays more rapidly than the normal and the tangential it is neglected. For illumination by a plane polarized wave; $U_0 = \exp(ikz+i\omega t)$.

$$U = \frac{S(\theta, \phi)}{ikr} \exp(ikr + i\omega t)$$

$$V = \frac{S(\theta, \phi)}{ikr} \exp(-ikr + ikz) U_0$$

Therefore the scattered field components in general are given by [7];

$$\begin{bmatrix} E_{11} \\ E_r \end{bmatrix} = \begin{bmatrix} S_2(\theta, \phi) & S_3(\theta, \phi) \\ S_4(\theta, \phi) & S_1(\theta, \phi) \end{bmatrix} \begin{bmatrix} E_{11}^0 \\ E_r^0 \end{bmatrix} \frac{\exp(-ikr+i\omega t)}{ikr}$$

Where E_{11} and E_r are the parallel and the perpendicular field components with respect to the plane of observation. For spherical particles, $S_3(\theta, \phi) = 0 = S_4(\theta, \phi)$ and the scattered intensities are given by:

$$I_r = \frac{i_1}{k^2 r^2} I_0$$

$$I_{ll} = \frac{i_2}{k^2 r^2} I_0$$

For any state of polarization the scattered intensity is:

$$I = \frac{i_1 + i_2}{2k^2 r^2} I_0 \dots (1-123)$$

where $i_1 = |S_1(\theta)|^2$ and $i_2 = |S_2(\theta)|^2$ are the basic Mie functions [8]. In the case of experimental measurement, a crosssection of scattering is very important which is given by:

$C_{sca} = \frac{1}{k^2} \int F(\theta, \phi) ds$; $F(\theta, \phi)$ being defined by the relation; $I = \frac{F(\theta, \phi)}{k^2 r^2} I_0$. The scattering crosssection is the crosssection of scattered energy [8]. That is the total energy scattered in all directions is equal to the energy of the incident wave falling on the area C_{sca} . In addition the efficiency factor given by $Q_{sca} = C_{sca}/G$, where $G = \pi a^2$, is of physically important value. It can be expressed as [7]:

$$Q_{sca} = \frac{2}{X^2} \sum_{n=1}^{\infty} (2n+1) (a_n^2 + b_n^2) \dots (1-124)$$

$$X = \frac{2\pi a}{\lambda}$$

The other quantity of interest in the scattering problem is the extinction coefficient; $\gamma_{ext} = \gamma_{sca} + \gamma_{abs}$. For a medium containing polydisperse system of N-particles, the intensity of the proceeding wave decreases by a factor of $\exp(-\gamma l)$ in a distance l .

For this system the extinction coefficient is given by;

$$\gamma = \int_0^{\infty} Q_{\text{ext}} \phi(a) G da \dots (1-125)$$

Hence physically measurable quantity, contains the distribution function $\phi(a) G da$. Using equation (1-124) the more general form of the scattered intensity obtained is;

$$I = \int_0^{\infty} \frac{F(\theta, \phi)}{K^2 (r-a)^2} \phi(a) G(a) I_0 da \dots (1-126)$$

Where $G(a)$ is the shape factor and for spherical particles; $G(a) = \pi a^2$ where a is the size of particle and r is the distance from the system to the point of observation. Therefore the appearance of the distribution function in the measurable quantities is meant to the determination of the SDF $\phi(a)$.

1.2 SCATTERED LIGHT AS A MEANS OF A MEASURE OF SIZE

A remarkable efforts have been done to determine size (SD) from the measurements of scattered intensities. Assuming that $(a)/m-a/\ll 1$, where $k = \frac{2\pi}{\lambda}$ and $a \sim r \ll \lambda$ as indicated in [7].

$$I = \frac{I_0 k^2 V^2 (m-1)}{r^2 8\pi^2} /R(\theta, \phi)/^2 (1+\cos^2 \theta),$$

where I is the scattered intensity and $R(\theta, \phi) = \frac{1}{V} \int \exp(i\delta) dV$ for $\delta = kr \cdot (\vec{m} - \vec{n})$, $\vec{m}$ and $\vec{n}$ being directions in the incident and the scattered light. It was also shown that $\delta = 2kb \sin(\theta/2)$ where δ is the phase angle and b is the bisectrix [7]

$$\begin{pmatrix} S_1(\theta) \\ S_2(\theta) \end{pmatrix} = \frac{ik^3 (m-1)}{2\pi} \int \exp(i\delta) dV \begin{pmatrix} 1 \\ \cos \theta \end{pmatrix}$$

where dV is the volume element of the system. In the limiting case $r \ll \lambda$ and for plane polarized incident light (Rayleigh) the intensity scattered at an angle θ from the incident direction is;

$$I_{\theta} = \frac{8\pi^2}{r^2} \frac{m^2 - 1}{m^2 + 2} \frac{V^2}{\lambda^4} \sin^2 \theta \dots (1-21)$$

In principle equation (1-21) can be used to determine the size of spherical particles. Nevertheless; the size of a particle of uniform droplet size can be obtained by measuring I_{θ} at angles varying from a forward to a backward direction and integrating over the spherical surface [9]. Extensive methods of size determination from scattering principles have also been discussed in [10], for particles having smaller size than the wave length used.

For sphere of radius "a" a function $G(u)$ is defined [11];

$$G(U) = \frac{1}{V} \int_{-1}^1 e^{-izu} \pi a^2 (1-z^2) dz;$$

where $U = \frac{4\pi a}{\lambda} \sin \frac{\theta}{2}.$

Using the condition, that at first minimum of two different size a_1 and a_2 ;

$$a_1^2 G(U_1) = a_2^2 G(U_2).$$

The values of $G(U)$ are tabulated for different U . For two different angles θ_1, θ_2 corresponding to two different wave length λ_1 and λ_2 . U has been evaluated interms of a_1 and a_2 . The size was then determined from the intersections of the two

graphs of $G_1(U)$ and $G_2(U)$ versus "a" [12], as given in /1/. In (1-21) r is the distance from the sphere to the point of observation and θ is the angle between the direction of propagation of the scattered and the incident light. For unpolarized incident light the intensity scattered in the direction θ is given by

$$I_{\theta} = \frac{9\pi^2}{2r^2} \cdot \frac{m^2-1}{m^2+2} \cdot \frac{V^2}{\lambda^2} (1+\cos^2\theta) \dots (1-22)$$

Equation (1-22) displays the fact that the scattered light by sphere of any size is composed of scattered light from two incoherent plane of polarization which are mutually perpendicular. In this case the " $\cos^2\theta$ " term, in equation (1-22), gives relative intensity of the scattered component whose electric vector lies in the plane defined by the incident beam and the observed scattered beam. The term unity in equation (1-22), represents the scattered component perpendicular to the plane of observation, when multiplied by the factor. For spheres of any size the general form of equation (1-22) is given in [7].

$$I = \frac{\lambda^2}{8\pi^2} (i_1 + i_2)$$

which is equivalent to equation (1-123). The complexity of the different methods arises in the case of polydisperse systems and failur to identify the property that clearly characterizes size, despite these; different approaches have still been excuted to get to a consistent one.

CHAPTER 2

RETROSPECTIVE METHODS

2.1 MICROPHOTOGRAPHIC METHOD (MPGM)

This method is the most simple and precise one. One of the typical (MPGM) for dispersion particles has been discussed in [13]. In this method a photographic image of colloidal specimen placed on a plane has been magnified (2500-10000) times. Depending on the dimensions of particles; 1000 to 2000 particles were compared with the area of circles drawn on a transparent glass. The method of correcting coefficient was suggested [12], because of the wide range of the values of the areas of particles and circles. A more convenient method was suggested [8] using a scanning beam. A beam of light crosses the particles straightly, the image is transformed into a series of electric impulses. The time of each impulse corresponds to the length of the particle which is viable to define the density of distribution function $\phi(x)$ of the electric impulse in accordance with the dimension of the particle.

For monodisperse particles with radius R_0 , the probability $d\omega$, that the beam of light will cross the particle in the interval $(y, y+dy)$ is; $d\omega = dy/R_0 \dots (2.11)$. But for circular particle $x^2+y^2 = R_0^2$ holds and;

$$d\omega = \frac{x dx}{R_0 \sqrt{R_0^2 - x^2}} \dots (2.12)$$

Because of the relation; $\phi(x) dx = d\omega \dots (2.13)$ and from equation (2.12), the SDF for monodisperse system will be;

$$\phi(x) = \frac{x}{R\sqrt{R^2-x^2}} \dots (2.14)$$

Similarly for polydisperse particles the SDF depends upon the dimensions of the particles. In this case the general form of the SDF can be deduced from equation (2.14) and is;

$$\phi(x) = \text{ReI} \int_0^\infty \frac{F(\sqrt{x^2+y^2})xdR}{y\sqrt{x^2+y^2}} \dots (2.15)$$

Where $F(R)$ is a correcting factor to a circular shape. Equation (2.15) can be rewritten as;

$$\frac{\phi(x)}{x} = \int_0^\infty \frac{F(\sqrt{x^2+y^2})xdR}{x^2+y^2} \dots (2.16)$$

Denoting $\frac{\phi(x)}{x} = H(x)$ and $\frac{F(\sqrt{x^2+y^2})}{x} = \psi(x);$

(2.16) can be written as;

$$H(x) = \int_0^\infty \psi(x^2+y^2)dy \dots (2.17)$$

Where in many cases of practical distribution the function $H(x)$, can be rearranged to have the form;

$$H(x) = e^{-\alpha x^2} \sum_{m=1}^\infty a_m x^{2m} \dots (2.18)$$

For the type of the function $H(x)$, equation (2.17) has the form;

$$F(R) = e^{-\alpha R^2} \{2a_0(\alpha/\pi - \frac{a_1}{2\pi})R^2 + 2a_1(\alpha/\pi)R^4\}$$

Which is the SDF for MPGM. Obviously the validity of this method depends upon the functional relation given in equation (2.18). In search of satisfactory method, an emperical approach based upon

the spectrophotometric measurement of reflected light was divided [12].

2.2 DETERMINATION OF SDF BY LIGHT SCATTERING AT THE SMALL ANGLE

This method is based upon the measurement of the scattering indicatrix [14] at the small angles. To solve this problem three assumptions were made. These are;

1. Particles are spherical in shape,
2. They are homogeneous,
3. Light is scattered only once.

As discussed in the preceeding section, this method has its foundation on the principle of electromagnetic diffraction by small monodisperse spheres, which leads to the expression of SDF for monodisperse indicatrix. The scattered intensity is;

$$I(\rho, \beta) = \frac{2}{\rho^2} J_1^2(\rho, \beta) \quad (2-21)$$

when $\rho \rightarrow \infty$, $\beta \rightarrow 0$, where ρ is defined from equation (2.14) as it is

where I = Intensity of scattered light;

β = Scattering angle;

a = Radius of the particle; conventionally application

λ = Wave length;

$\rho = \frac{2\pi a}{\lambda}$, and $J_1(\rho, \beta)$, bessel function of the first kind.

Equation (2.21) is valid if $\rho > 30$. In the case of polydisperse system the intensity of the scattered light can be written as

the sum of the intensities scattered.

$$I_{\Sigma}(\beta) = \sum_N I_N(\rho, \beta)$$

for all particles for small scattering angle [15]

$$I(\beta) = \int_0^{\infty} I(\rho, \beta) \phi(\rho) d\rho \dots (2.22)$$

Equation (2.22) can be written as;

$$I_{\Sigma}(\beta) = \int_0^{\rho} I(\rho, \beta) \phi(\rho) d\rho + \int_{\rho}^{\infty} I(\rho, \beta) \phi(\rho) d\rho \dots (2.23)$$

Here; ρ is the value at which equation (2.21) gives the most exact value. For large particles, the first term is much smaller than the second term, in that case the equation reduces to;

$$I(\beta) = C \int_{\rho}^{\infty} \frac{\phi(\rho) \rho^2 J_1^2(\rho, \beta)}{\beta^2} d\rho \dots (2.24)$$

The SDF, $\phi(\rho)$, can be determined from equation (2.24), as it is a homogeneous integral equation. In this method it is possible to obtain the result more or less quickly. Hence for rapidly fluctuating process, the method is more conveniently applicable. But the draw-back of this method is in its considerable errors for values obtained. For large angle of measurement, the error is some (50-70)%; due to the fact that $I_{\Sigma}(\beta)$ decreases very quickly when the angle increases. This method is rather widely used in the investigation of aerosols.

2.3 DEPOLARIZATION

This method has its foundation in the Mie Scattering theory for spherical particles. The light scattered at the angle $\theta = \pi/2$ is only partly polarized. The degree of its polarization depends upon the size and shape of the scattering particles. Depolarization relation is determined by;

$$\sigma = \frac{I_{11}}{I_{1r}} \dots (2.31)$$

where

$$I_{11} \text{ \& } I_{1r}$$

are the intensities of scattered light at $\theta = \pi/2$, in the perpendicular and parallel planes respectively. The intensities of the light scattered in unite angle which correspond to particles of radius r are given by;

$$I_{11} = \frac{\lambda^2}{4\pi^2} i_{11}(\alpha) \dots (2.32)$$

and $I_{1r} = \frac{\lambda^2}{4\pi^2} i_{1r}(\alpha) \dots (2.33)$

where

$$\alpha = \frac{2\pi r}{\lambda}; \quad i_{11} \text{ \& } i_{1r}$$

are basic Mie functions. Corresponding to the scattered light

at $\theta = \pi/2$, i_{11} and i_{1r} depend on the relative refractive index

$$n = \frac{n_p}{n}; \text{ where}$$

n_p is refractive index of particles and

n is refractive index of the medium.

For polydisperse system we have

$$I_{11} = \int_0^\infty I_{11}^0 C\phi(r) dr \dots (2.34)$$

$$I_{1r} = \int_0^\infty I_{1r}^0 C\phi(r) dr \dots (2.35)$$

Here $\phi(r)$, is the distribution function in size, C is the normalization constant.

The SDF assumed [16] has the form;

$$\phi(r) = \begin{cases} (r-r_0) \exp(-(\frac{r-r_0}{S})^3); & \text{for } r \geq r_0 \\ 0; & \text{for } r < r_0 \end{cases} \dots (2.36)$$

Where r_0 and S are parameters of distribution function. r_0 is the smallest radius of particles and S is the modal radius. He by determining r_0 and s it is possible to find $\phi(r)$. Taking in account equation (2.31), (2.32), (2.33) and (2.36) we get

$$\sigma = \frac{F_{11}(p,q)}{F_{1r}(p,q)} \dots (2.37)$$

where

$$p = \frac{2\pi r_0}{\lambda}$$

and

$$q = \frac{2\pi s}{\lambda};$$

Practically it is possible to find σ . Plotting the theoretical value of σ_{exp} , p and q can be found from the coincidence of the two curves. As a result r_0 and s can be determined for the determination of $\phi(r)$ as discussed in [16].

2.4 NEPHELOMETER

The technique, as in the case of other methods, is based on the ability of colloidal systems to scatter light. By determining the intensity of light scattered by a system it is possible to determine the size of the particles and SDF. According to Mie theory;

$$\tau/c = \frac{3\pi m}{100\lambda d^3} \sum_j \frac{a_j^2/b_j^2}{2j+1} \dots (2.41)$$

Where τ is the scattering coefficient; C ratio of volume of particles to total volume of colloide, n is the refractive index. For a given λ ,

$$\alpha = \frac{\pi n d}{\lambda}$$

where " d ," is the diameter of particles. It is assumed that all particles are sphere of equal diameter. a_j and b_j are functions of α and

$$m = \frac{n_p}{n}$$

As the thickness of the medium through which light is propagating increases the intensity of light decreases and this can be stated in equation form as;

$$-\frac{dI}{I dx} = \tau \dots (2.42)$$

where I is the intensity of the incident light and " dx ," is the thickness of the layer; the value of τ/C is calculated theoretically and the result is given in [17] for $\lambda = 5461 \text{Å}$. For this value of wave length of the light the change of τ/c for small change of m is very sharp in accordance with the relation (2.41). For smoothening of this sharpness a logarithmic operation has been used in order to interpolate between consecutive tabular values. Hence;

$$\log \frac{\tau}{C} = \log \frac{n}{C} + I_0(\alpha, m) + S_0(\alpha, m).$$

This gives

$$\log \frac{\tau}{C} = \log \frac{n}{C} + I_0(\alpha, m) \log(m-1) \dots (2.43)$$

It was shown, [18], that the function is approximately linear to $\log(m-1)$, when τ is maximum, and also $\log \frac{\tau}{C} \propto \log \lambda$. For the case of maximum λ , in the visible region, the slope " s " of the curve $\frac{\tau}{C}$ versus λ , will be; [19]

$$S = - \left(\frac{\partial \log \tau}{\partial \log \lambda} \right) \Big|_d$$

$$\begin{aligned} \bar{S} = & - \left(\frac{\partial \log \tau}{\partial \log n} \right) \Big|_{\alpha, m} \frac{d \log (n/\lambda)}{d \log \lambda} - \left(\frac{\partial \log \tau}{\partial \log \lambda} \right) \Big|_{m, \frac{n}{\lambda}} \left(\frac{\partial \log \alpha}{\partial \log \lambda} \right) \Big|_{\bar{d}} \\ & - \left(\frac{\partial \log \tau}{\partial \log (m-1)} \right) \Big|_{d, \frac{n}{\lambda}} \left(\frac{d \log (m-1)}{d \log \lambda} \right) \end{aligned}$$

From equation (2.43), the equation obtained is;

$$\left(\frac{\partial (\log \alpha)}{\partial \log \lambda} \right) \Big|_d = \left(\frac{d \log n}{d \log \lambda} \right) - 1$$

and
$$\left(\frac{\partial \log}{\partial \log} \right) \Big|_{m, \frac{n}{\lambda}} = \left(\frac{\partial \log \tau}{\partial \log d} \right) \Big|_{m, \frac{n}{\lambda}} ;$$

the resulting equation hence is;

$$S = - \left[1 - \left(\frac{d \log n}{d \log \lambda} \right) \right] \left[1 + \left(\frac{\partial \log \tau}{\partial \log d} \right) \Big|_{m, \frac{n}{\lambda}} \right] - \frac{\partial \log \tau}{\partial \log (m-1)} \Big|_{d, \frac{n}{\lambda}} \frac{d \log (m-1)}{d \log \lambda}$$

The values of $\frac{d \log (m-1)}{d \log \lambda}$ and $\frac{d \log n}{d \log \lambda}$, can be estimated from theoretical tables or from the plot of the function τ/c . The other two terms are determined experimentally, which are necessary in finding the values of τ or τ/C , **sometimes** it is more convenient to use the value,

$$\frac{S\tau}{c} = - \left(\frac{\partial (\tau/c)}{\partial \log \tau} \right) \Big|_d$$

where as, for polydisperse system

$$\tau = \sum_i C_i \left(\frac{\tau}{c} \right)_i$$

The light scattering coefficient for such a system can be written in integral form as;

$$\left(\frac{\tau}{C}\right)\Big|_0 = \int_0^{\infty} \frac{\tau}{C} F(d) \delta d \quad \dots \quad (2.45)$$

Where $F(d)$ is the distribution function of particles in size.

We can write:

$$\begin{aligned} \left(\frac{S\tau}{C}\right)\Big|_0 &= \int_0^{\infty} \left(\frac{\partial(\tau/C)}{\partial \log \lambda}\right)\Big|_d F(d) \delta d \\ &= \int_0^{\infty} (S\tau/C) F(d) \delta \bar{d} \quad \dots \quad (2.46) \end{aligned}$$

τ/c and $S\tau/c$ in equations (2.45) and (2.46) can be calculated and give us the possibility to write $F(d)$ as;

$$\left(\frac{\tau}{C}\right)\Big|_0 = \sum_{i=1}^n \left(\frac{\tau}{C}\right)\Big|_i \int_0^{\infty} F(d) \delta d \quad \dots \quad (2.47)$$

$$\left(\frac{S\tau}{C}\right)\Big|_0 = \sum_{i=1}^n \left(\frac{S\tau}{C}\right)\Big|_i \int_0^{\infty} F(d) \delta d \quad \dots \quad (2.48)$$

In case of logarithmic normal law of distribution equation (2.47) and (2.48) can be rewritten as;

$$\begin{aligned} \left(\frac{\tau}{C}\right)\Big|_0 &= \sum_{i=1}^n f_i \left(\frac{\tau}{C}\right)\Big|_i, \\ \left(\frac{S\tau}{C}\right)\Big|_0 &= \sum_{i=1}^n f_i \left(\frac{S\tau}{C}\right)\Big|_i \end{aligned}$$

where f_i is given by

$$f_i = \frac{1}{\sqrt{2\pi}} \int_{t_i - \Delta t_i}^{t_i} e^{-\frac{1}{2}t^2} dt;$$

where

$$t = \frac{\log d - \log \bar{d}}{\log \sigma}$$

for an average diameter $\bar{d}$ of particles and σ characterizes the half width of the curve [27], [20].

2.5 METHOD OF LIGHT SCATTERING FOR RIGID-STICK PARTICLES

This method was developed in [20]. For monodisperse system the relative alteration of the intensity can be described as:

$$a_t = a_0 e^{-t/\tau} \dots (2.51)$$

where

$$a = \frac{I_t - I_0}{I_0}$$

and $a|_{t=0} = a_0$; I_t and I_0 are intensities of the scattered light by one particle, when the electric field is switched on and switched off respectively. Then

$$\tau = \frac{1}{D} \dots (2.52)$$

is the time in which

$$a_t = \frac{a_0}{e}$$

and is called the relaxation time. D is the rotary diffusion coefficient. For polydisperse system of small concentration;

$$\frac{a_t}{a_0} = \frac{\sum_{i=1}^n \alpha_{oi} e^{-\frac{t}{\tau_i}}}{\sum_{i=1}^n \alpha_{oi}} \dots (2.53)$$

The value of optical signal for such particles, cylinders with different length and equal diameter, can be written as:

$$a = \frac{\sum_{i=1}^n n_i l_i^2 I_{oi} a_i}{\sum_{i=1}^n n_i l_i^2 I_{oi}} \dots (2.54)$$

Where n_i is quantity of particles and l_i = length. Taking into account equations [2.53] and [2.54] and

$$\alpha = \frac{I_E - I_0}{I_0}$$

we obtain;

$$\frac{\sum_i n_i l_i^2 (I_E - I_0) e^{-\frac{t}{\tau_i}}}{\sum_i n_i l_i^2 (I_E - I_0)_i} = \frac{\alpha_t}{\alpha_0} \dots (2.55)$$

In order to determine the SDF, n_i and l_i have to be determined. In this respect, the experimental curve of the relaxation time of the particle after switching off the electric field has to be used. Experimentally it is possible to determine [21] graphical curves at the separate parts. It means the real experimental curve is the combination or sum of the separate or individual curves which correspond to define group of particles. Thus family of exponential curves for real experimental one can be written as;

$$\frac{\alpha_t}{\alpha_0} = \frac{n}{\sum_{i=1}^n} e^{-\frac{t-t_{i0}}{\tau_i}} \dots (2.56)$$

comparing equation [2.55] and [2.56], the result is:

$$n_i = \frac{\exp(\frac{t_{i0}}{\tau_i}) C}{l_i^2 (1 - I_{oi})} \dots (2.57)$$

where

$$C = \frac{1}{\sum_{i=1}^n n_i l_i^2 (I_E - I_0)}$$

Equation [2.57] gives the final result; for each length l_i the respective n_i can be found and the SDF can be plotted. $\tau_{i0}, \tau_i, I_{oi}$ are calculated by comparing experimental and theoretical curves in accordance with the method given by the author of this section [21].

2.6 ELECTROOPTICAL DIFFUSION METHOD FOR ESTIMATING COLLOIDAL PARTICLES SDF WITH THEIR ALLOWANCE OF LIGHT ATTENUATION

This was suggested and developed in [22]. The theory is based upon the electrooptical properties of particles, there by giving different approach to SDF. The value of the optical effect associated with the relaxation time of the colloidal particles orientation after the removal of the rotating $\vec{E}$ -field is assumed to be related with $\phi(r)$ through an integral equation. The stated problem is of none-correcting class (choice of parameters). A more detailed method of solution has been suggested [22]. The polarized light was send through colloid solution, which contains particles oriented by the rotating field. After switching off the field the rotary diffusion motion of particles begins from saturation state. Hence the intensity of the polarized light will have different values. The optical effect $N(t)$ is given by

$$N(t) = \frac{\Delta k}{L} \int_0^\pi (3 \cos^2 \theta - 1) F(\theta, t) \sin \theta d\theta \dots (2.61)$$

where

$$N(t) = \frac{I_{1r}(t)}{I_{11}(t)} \dots (2.62)$$

$I_{1r}(t)$ and $I_{11}(t)$ are light intensities crossing through colloid which are polarized perpendicular and parallel to the electric field respectively. $\Delta k = k_{11}(r) - k_{1r}(r)$ is the difference of extinction coefficient of particles of radius r along and across the long and short axes respectively. $F(\theta, t)$ is a function of the angular distribution of the axis of particles with respect to the direction of the electric field.

It can be shown that

$$N(t) = \Delta k (1 - 3/2 \overline{\sin^2 \theta}) \dots (2.63)$$

where $\overline{\sin^2 \theta} = \int_0^\pi \sin^2 \theta F(\theta, t) \sin \theta d\theta;$

If $t = 0$ and all particles were oriented completely then

$$\overline{\sin^2 \theta} = \frac{2}{3} (1 - e^{-6Dt}) \dots (2.64)$$

as given in [10]. Where D is the rotary diffusion coefficient, solving equation [2.63] and [2.64] $N(t)$ will be; $N(t) = \Delta k e^{-6Dt}$ [2.65]. This equation can be used for polydisperse system and the optical effect is;

$$N(t) = \int_0^a f(r) e^{-6D(r)t} dr \dots (2.66)$$

Where $f(r)$ is the SDF. $N(t)$ can be obtained experimentally and $f(r)$ has to be found in Tichonov's space W_2^1 of regular function with their metrix;

$$||f_1 - f_2|| = \int_0^a [(f_1 - f_2)^2 + (\frac{df_1}{dr_1} - \frac{df_2}{dr_2})^2]^{1/2} dr \dots (2.67)$$

The functions f_1 and f_2 are the elements of the space W_2^1 [23]. Hence by this approach it is possible to solve the problem by removing the none-correct class. In the conditionally correct class using the method of regularization for solving equation (2.66), the authors have found;

$$b(r) = \int_0^a K(r, \rho) f(\rho) d\rho - \alpha [\frac{d^2 f(r)}{dr^2} - f(r)] \dots (2.68)$$

where $K(r, \rho) = \frac{1 - e^{6[D(r) + tD(\rho)]t_0}}{6[D(r) + D(\rho)]}$

is the kernel of the integral equation. Also $b(r)$ reduces to

$$b = \int_0^{t_0} N(t) e^{-6D(r)t} dt$$

α is the parameter which is calculated from equation;

$$(\delta N)^2 = \int_0^{t_0} \left[\int_0^a e^{-6D(r)t} f^\alpha(r) dr - N(t) \right]^2 dt \dots (2.69)$$

Where $(\delta N)^2$ is experimental error, t_0 is the interval of time $[0, t_0]$ which determines time of measurement of $N(t)$.

2.7 LASER DIFFRACTION SYSTEM METHOD [24]

In this method it was pointed out that SDF to be determined using superimposed fraunhofer diffraction pattern of moving particles on a focal plane of a lense. In addition, the obscuration of diffracted light intensity of slits by obstacles behind the slits was analyzed and the influence of particle numbers in a unite volume on diffracted light intensity was estimated.

A system of Linear equations were developed and used to determine particle's radius. The energy on the torifaces, having different radii, L_1, L_2, L_3 were obtained from;

$$L_1 = a_{11} W_1 / b_1 + a_{12} W_2 / b_2 + \dots + a_{1n} W_n / b_n$$

$$L_2 = a_{21} W_1 / b_1 + a_{22} W_2 / b_2 + \dots + a_{2n} W_n / b_n$$

.

.

$$L_n = a_{n1} W_1 / b_1 + a_{n2} W_2 / b_2 + \dots + a_{nn} W_n / b_n$$

where W_j 's are gross weights.

The light energies were detected using a photodetector, and

these values were substituted in the above equation to obtain particles diameter $b_1, b_2, \dots b_n$.

The draw back of this method is that the number of particles restrict the values of particle's diameter. It was pointed out that the method of average intensity minimized the influence of obstacles on the measured intensity.

2.8 DETERMINATION OF SDF FROM THE SCATTERING INDICATRIX IN THE PRESENCE OF SPECKLE [14], [25]

In this method the SDF determination was inferred from the angular distribution (ADF) of the scattered intensity. A laser was used as a light source for self-luminous flows of particles. A speckle was observed in the recording plane. For particles size: $r \gg \lambda$, small-angle light diffraction was used. The AD of the scattered intensity considered was;

$$I(\theta) = \frac{C}{\theta^2} \sum_{i=1}^N r_i^2 J_1^2(kr_i \theta) \dots (2.81)$$

Where $k = 2\pi/\lambda$; $J_1(x)$ is Bessel's function; C, constant. In the presence of speckle the intensity scattered becomes;

$$S(\theta) = I(\theta) + 2 \sum_{i=1}^{N-1} \sum_{j=i+1}^{N-1} I_i(\theta) I_j(\theta) \cos(R_i - R_j) \dots (2.82)$$

Where R_i is the optical path length from the particle to the recording point. The dependence of $S(\theta)$ on the random particle distribution was used as the starting point of determination of SDF. This dependance was taken as; $S(\theta) = I(\theta) [1 + \xi] \dots (2.83)$

Where ξ is a measure of perturbation. The type of SDF obtained is; $f(r) = \frac{\alpha^{\mu+1} r^{\mu} e^{-\alpha r}}{\Gamma(\mu+1)}$ where α and μ , are distribution parameters and $\Gamma(x)$ is the gamma function.

CHAPTER 3

Determination of Size Distribution Function (SDF)

by Electrooptical Method

3.1 Theoretical Formulation of the Method

When particles are placed in a rotating electric field (R-field) [22,26], they will be oriented by the field. This field is set into rotation by using a three phase voltage input. The oriented colloids will rotate with angular velocity $\hat{\omega}$ lagging by a certain angle α , constant in time, from the field. The experimental scheme is shown in Figure 1.

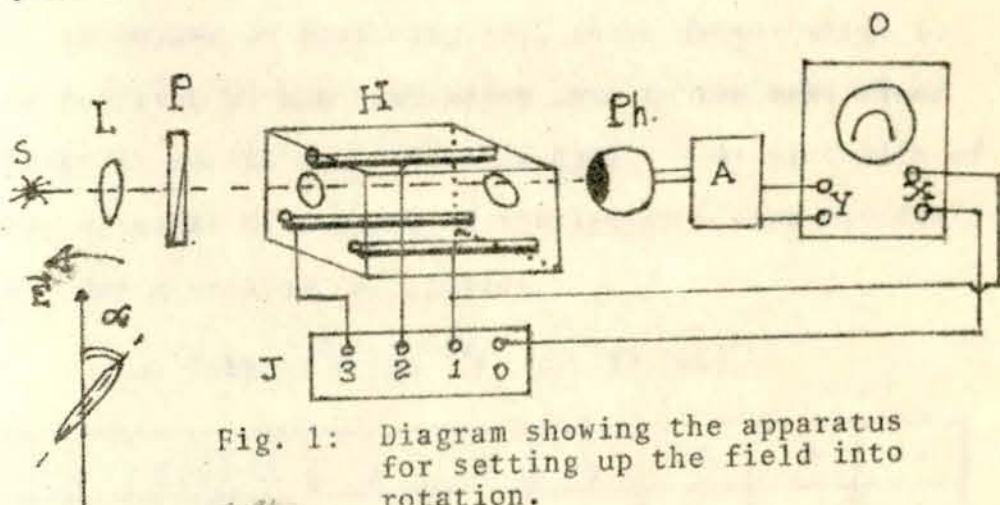


Fig. 1: Diagram showing the apparatus for setting up the field into rotation.

S, source; L, lens; P, polarizer; Ph, photomultiplier
O, oscillograph; A, amplifier;
H, solution with electrodes; J, generator.

Since the system of oriented colloidal particles possess dichroism, the solution is a kind of "Polaroid" and therefore acts as "analyzer". In the above optical scheme rotation of such analyzer with angular velocity $\hat{\omega}$, relative to the immovable polarizer "P" causes

sinusoidal modulation of the light with frequency 2ω . The depth of modulation depends on the perfection of the analyzer as a second polarizer. The modulated amplitude can be observed and measured on the screen of an oscillograph "O" after a preliminary amplification by the photomultiplier Ph. For particles having a permanent dipolmoment $\vec{\mu}$, the predominant factor of orientation in the R-field, the equation of motion is;

$$\mu E \sin \alpha = \eta V [P] \omega_F \dots (3.101)$$

where ω_F is the frequency of the R-field.

The left side of equation (3.101) represents the moment of the R-field, and the right side is due to the viscosity of the media. V , is volume of particle; $[P]$, shape factor which is related to P , ratio of the semi-major axis to the semi minor axis (Table 1) and is calculated in [27]. η is viscosity of the liquid; α , angle of lagging of the vector $\vec{\mu}$ from the field direction. For a prolate ellipsoid;

$$\mu = 3.8\mu_1 (V^2 P \cos)^{1/3} \dots (3.102)$$

P	1	1.5	2	3	4	5	6	8	10
[P]	6.0	7.5	9.0	14.0	20.1	27.7	35.8	56.1	80.5

Table 1: Calculated values of $[P]$ for a given value of P [27]

μ_1 in equation (3.102) is the sum of the dipolmoment of all water molecules capable of forming a single layer of 1 cm^2 and P is the ratio of the semi-minor axis to the semimajor of the ellipsoid. Since the molecules of water adsorb on the surfaces of colloidal particles, μ is purely surface property [28], and is associated with the angle

of inclination β of the water molecule dipole axis relative to the partial surface. If "l" is the long axis which corresponds to the direction $\vec{u}$, then;

$$l = \left[\left(\frac{3p^2}{4\pi} \right) V \right]^{1/3} \dots (3.103)$$

Obviously; $\sin \alpha = V[P] \eta \omega_F / \mu E \dots (3.104)$

after combining equations 3.102, 3.103 and 3.104, we have;

$$\sin \alpha = B(P) l \omega_F \dots (3.105);$$

where: $B(P) = \frac{\eta}{2.35 E l_1 (\cos \beta) P} \dots (3.106)$

For monoshape particles, the dependence of B on the parameter "P" is very weak, and is quasi-linear, (Fig. 2-a).

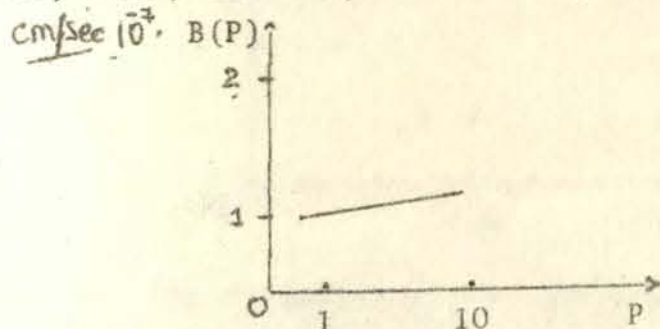


Fig.2 (a) Dependence of B on the parameter P for polydisperse monoshape particles.

Therefore,

$$\sin \alpha = B l \omega_F; \dots (3.107)$$

Hence from equation (3.107) it is clear that each kind of particle has its own angle α . If for a given ω_F , E and l are constants, the dependence of $\sin \alpha$, on ω_F is linear upto a critical frequency ω_{cr} , Fig. 2b.

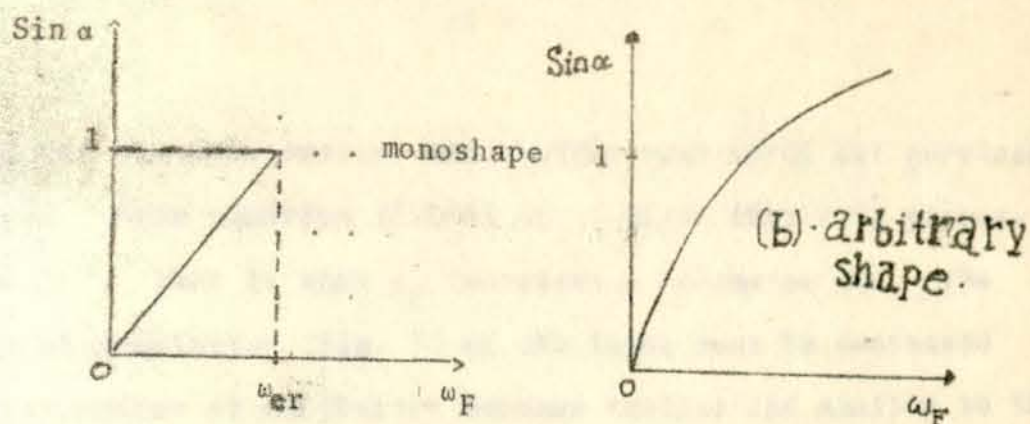


Fig. 2-b: Dependence of „Sin α ” on the field frequency, ω_F . The value of ω_F is increased only upto ω_{er} , which corresponds to the angle $\alpha = \pi/2$.

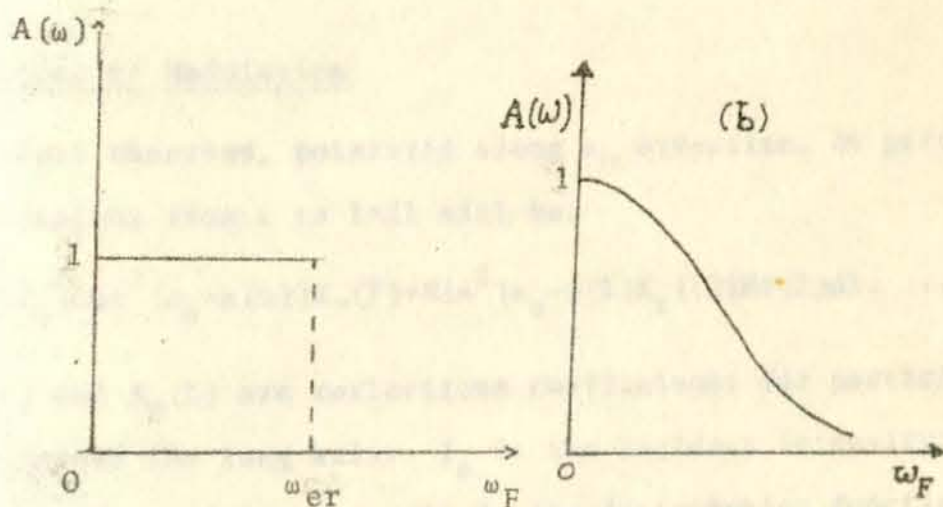


Fig. 3: The variation of the amplitude of modulation $A(\omega)$ with the field frequency ω_F . (a) For monoshape particles (b) For polydisperse system of particles of arbitrary shape.

Hence increasing of α could be possible upto $\pi/2$; after which the modulation of light would be stopped instantaneously. In the case of polydisperse colloid such picture is not possible. First the most big particles will stop its orientation when the angle α assumes the value $\pi/2$. If further ω_F is increased, the situation

will hold for the more smaller and smaller ones until all particles are exosted. From equation (3.104) it is clear that α is proportional to "1"; that is when ω_F increases α increases too. The amplitude of modulation (Fig. 3) of the light must be decreased because the number of mudulators becomes smaller and smaller as the frequency increases. Therefore, it is clear that experimentally measured value of $A(\omega)$ contains the real information concerning the distribution function in size.

3.2 Amplitude of Modulation

The light absorbed, polarized along α_0 direction, by particles having dimensions from 1 to $1+dl$ will be:

$$-dI(1) = I_0 \{ \cos^2(\alpha_0 - \alpha(1)) K_{||}(1) + \sin^2(\alpha_0 - \alpha(1)) K_{\perp}(1) \} N \phi(1) dl. \quad \dots (3.201)$$

where $K_{||}(1)$ and $K_{\perp}(1)$ are reflections coefficients for particles along and across the long axis. I_0 is the incident intensity; N , total number of particles and $\phi(1)$ is the distribution function in size. For particles, having arbitrary dimensions:

$$I(1_{cr}) = I_0 \int_0^{1_{cr}} \{ \cos^2(\alpha_0 - \alpha(1)) K_{||} + \sin^2(\alpha_0 - \alpha(1)) K_{\perp} \} N \phi(1) dl. \quad \dots (3.202)$$

where $1_{cr} = \frac{1}{B\omega_F} \dots (3.203)$. The value $I(1_{cr})$ has an extremum which corresponds to one of the axes of the colloidal polaroide.

At the extremum values:

$$\frac{dI(1)}{d\alpha_0} = 0 \quad \dots (3.204)$$

The extremum condition is specified by $\bar{\alpha}$, when $\bar{\alpha}$, satisfies the relation:

$$\tan(2\bar{\alpha}) = \frac{\int_0^{1_{cr}} f(l) \sin 2\alpha(l) dl}{\int_0^{1_{cr}} f(l) \cos 2\alpha(l) dl} \quad \dots (3.205)$$

The amplitude of the modulated polarized light, passing through the system of dichroitic particles is proportional to the difference of intensities of light at plane of polarization along and across the direction made by $\bar{\alpha}$ and is given by:

$$A(1_{cr}) = I_{\bar{\alpha}} - I_{\bar{\alpha}+90^\circ} \quad \dots (3.206)$$

where

$$I_{\bar{\alpha}}(1) = I_0 [1 - \int_0^{1_{cr}} K(1, \alpha) N \Phi(1) dl] \quad \dots (3.207)$$

$$\text{and } K(1, \alpha) = \cos^2(\bar{\alpha} - \alpha(1)) K_{||}(1) + \sin^2(\bar{\alpha} - \alpha(1)) K_{\perp}(1)$$

$$\text{Also, } I_{\bar{\alpha}+90^\circ}(1_{cr}) = I_0 [1 - \int_0^{1_{cr}} H(1, \alpha) N \Phi(1) dl] \quad \dots (3.208)$$

$$\text{Here } H(1, \alpha) = \sin^2(\bar{\alpha} - \alpha(1)) K_{||}(1) + \cos^2(\bar{\alpha} - \alpha(1)) K_{\perp}(1).$$

Consequently, the amplitude of modulation is equal to:

$$A(1_{cr}) = I_0 \int_0^{1_{cr}} \{ \cos^2(\bar{\alpha} - \alpha(1)) - \sin^2(\bar{\alpha} - \alpha(1)) \} \Delta K N \Phi(1) dl \quad \dots (3.209)$$

Representing, $f(1) = N \Delta K(1) \Phi(1)$; $\Delta K(1) = K_{||}(1) - K_{\perp}(1)$;

equation (3.209) can be rewritten as;

$$A(1_{cr}) = I_0 \int_0^{1_{cr}} f(1) dl - 2 I_0 \int_0^{1_{cr}} \sin^2(\bar{\alpha} - \alpha(1)) f(1) dl \quad \dots (3.210)$$

Differentiating equation (3.210) with respect to 1_{cr} , we obtain;

$$\frac{dA(1_{cr})}{dl_{cr}} = I_0 \{ f(1_{cr}) - F(1_{cr}) \} \quad \dots (3.211)$$

where $F(l_{cr})$ is the derivative of the second integral in equation (3.210). For most real models the value of the term $F(l_{cr})$, has been found out to be (3 to 4)% of $f(l_{cr})$, and is neglected in comparison with $f(l_{cr})$. Equation (3.211) in this case becomes;

$$\frac{dA(l_{cr})}{dl_{cr}} = I_0 f(l_{cr}), \dots (3.212).$$

Using equation (3.203), the resulting equation will therefore be;

$$\frac{dA(l_{cr})}{dl_{cr}} = \frac{dA(1/\omega_F)}{d(1/\omega_F)} = I_0 f(l_{cr}) \dots (3.213)$$

3.3 Determination of l_{cr}

In the qualitative analysis of the preceeding section, it is seen that both the amplitude of modulation and the slope of the graph of $A(1/\omega_F)$, versus ω_F are dependent on the critical length l_{cr} of each particle. In order to determine l_{cr} ; B, has to be known priorly. For this two ways have been employed. The first method is, "Electrooptical τ -meter method [29]. In this case it is necessary to measure the relaxation time τ , of disorientation of particles and to find the average volume of particles;

$$\bar{V} = \frac{6kT\tau}{[P]\eta} \dots (3.301);$$

where T is temprature; k , Boltzmann's Constant and η is the viscosity of the liquid used. For monoshape particles;

$$\overline{l^3} = \frac{9kT\tau P^2}{2\pi\eta[P]} \dots (3.302),$$

where l is the radius of an ellipsoid. On the other hand equation (3.107), gives;

$$\overline{l^3} = \frac{1}{B\omega_F}^3.$$

By the help of equation (3.213) the expression obtained is;

$$\overline{l^3} = \frac{1}{B\omega_F}^3 = \frac{\frac{1}{B^3} \int_0^\infty \left(\frac{1}{\omega_F}\right)^3 \frac{dA\left(\frac{1}{\omega_F}\right)}{d\left(\frac{1}{\omega_F}\right)} d\left(\frac{1}{\omega_F}\right)}{\int_0^\infty \frac{dA\left(\frac{1}{\omega_F}\right)}{d\left(\frac{1}{\omega_F}\right)} d\left(\frac{1}{\omega_F}\right)} \dots (3.303)$$

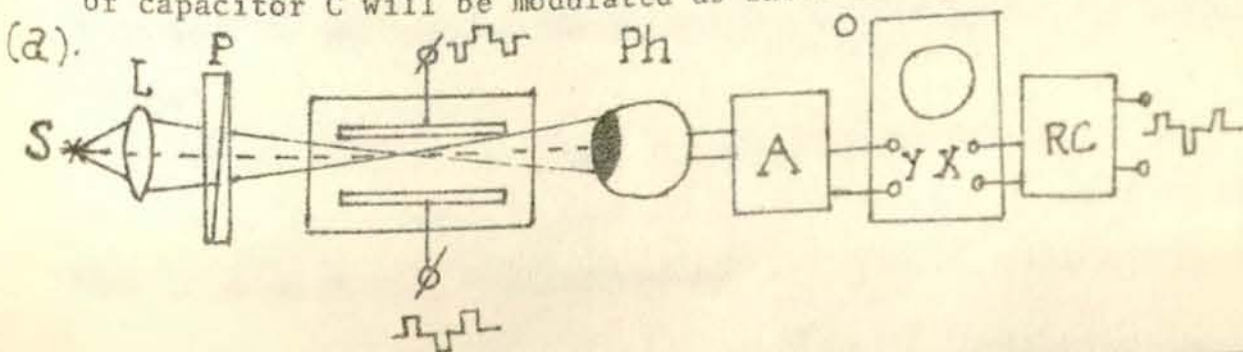
Comparing equation (3.301) and (3.303), B is given by;

$$B = \left[\frac{2\pi\eta[P]}{9kT\tau Y} \right]^{1/3} \dots (3.305)$$

where Y is the value of the integral in equation (3.303). Since all values in equation (3.305) can be measured experimentally, B can be determined from experiment.

3.4 Determination of τ by Electrooptical τ -meter

When rectangular electric pulses are applied to a colloidal solution; (Fig. 4a), the polarized light passing through the plate of capacitor C will be modulated as shown in Fig. 4b.



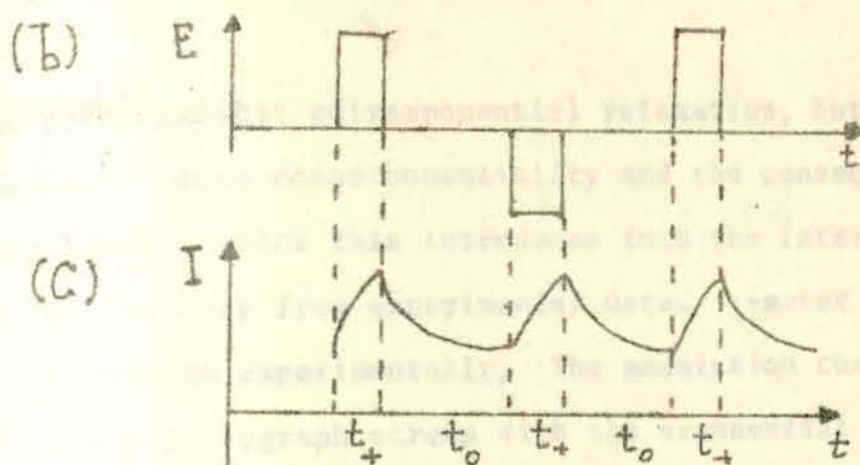


Fig. 4: Apparatus and the modulated impulse for investigating the orientation - relaxation curve and for measuring τ , with τ -meter.

S, source; L, lense; P, polarizer, Ph, photo-multiplier; A, amplifier; O, oscillograph; (a) diagram of τ -meter, (b) the form of rectangular electric pulses, (c) modulation curve for light passing through a colloidal solution subjected to the rectangular electric pulses. I -intensity of transmitted light t_+ and t_- periods of field action (orientation periods); t_0 -Interruption period (relaxation time).

During the time of the pulse action (t_+ or t_- Fig. b and c), the particles are oriented by the field. Relaxation occurs in the intervals between t_0 . During the period t_0 , the intensity I of the polarized light decreases or increases in accordance with the expressions; $I = I_0 \exp(-t/\tau)$ or $I = I_0[1 - \exp(-t/\tau)]$ (3.401) depending on the sign of the electrooptical effect for the given colloid.

Formula (3.401) is valid for monodisperse colloid of the same shape and therefore gives a sign +. The corresponding relaxation is described as monoexponential one. Real solution, in

general, exhibit polyexponential relaxation, but the degree of deviation from monoexponentiality and the consequent degree of uncertainty which this introduces into the interpolation can be estimated only from experimental data. τ -meter method decides this problem experimentally. The modulation curves are observed on the oscillograph screen with the exponential time scale, triggered by the rectangular electric pulses passing through the integrated RC-circuit. When the selected value of RC becomes equal to τ , for the same exponential law, a straight line appears on the screen of the oscillograph. If the relaxation curve cannot be linearized, the same exponential law will not hold. If, most or at least a considerable particles relaxe in accordance with the same exponential law, the relaxation curve will be quasi-linear (by selection of RC) over a considerable region.

The other method of determining B has been described in [28]. It is based upon the values of ν_1 , β and P through equation (3.306).

3.5 Method of Determination of SDF

From equation (3.201), it is possible to find an expression containing the SDF $\phi(r)$, where for spherical particles

$$r = \left(\frac{3V}{4\pi}\right)^{1/3},$$

and V is the volume of particles. For particles approximating to a prolate ellipsoidal revolution, 1- can be given by;

$$1 = \left[\frac{3P^2}{4\pi} V \right]^{1/3} = \left(\frac{3P^2}{4\pi} \right)^{1/3} = P^{2/3} r.$$

Also for spheres $P = 1$ and $r = \frac{1}{B\omega_F} \dots (3.501)$

The expression which involves the distribution function in size, $\phi(r)$, follows from equation (3.212), which is;

$$\frac{dA(1/\omega_F)}{d(1/\omega_F)} = I_0 N \Delta K \phi(r) \dots (3.502)$$

The left side of equation (3.502) can be determined experimentally and I_0 is the measured intensity. Thus we have three unknowns, N , $\Delta K(r)$ and $\phi(r)$. Two more equations are required in order to obtain $\phi(r)$ explicitly. In general the relational dependence of the three unknowns can be written as;

$$\begin{aligned} \phi_1(N_1, I_0 \Delta K(r), \phi(r)) &= 0 \\ \phi_2(N_2, I_0 \Delta K(r), \phi(r)) &= 0 \end{aligned} \quad \dots (3.503)$$

Therefore, equations 3.502 and 3.503 give;

$$\begin{aligned} \phi_1(N_1, I_0 \Delta K(r), \phi(r)) &= 0 \\ \phi_2(N_2, I_0 \Delta K(r), \phi(r)) &= 0 \end{aligned} \quad \dots (3.504)$$

$$\frac{dA(1/\omega_F)}{d(1/\omega_F)} = I_0 N \Delta K(r) \phi(r)$$

To obtain dependences (3.504-1) and (3.504-2) it is possible to perturb the distribution function of an equilibrium state, in a certain volume V_1 (Fig. 5).

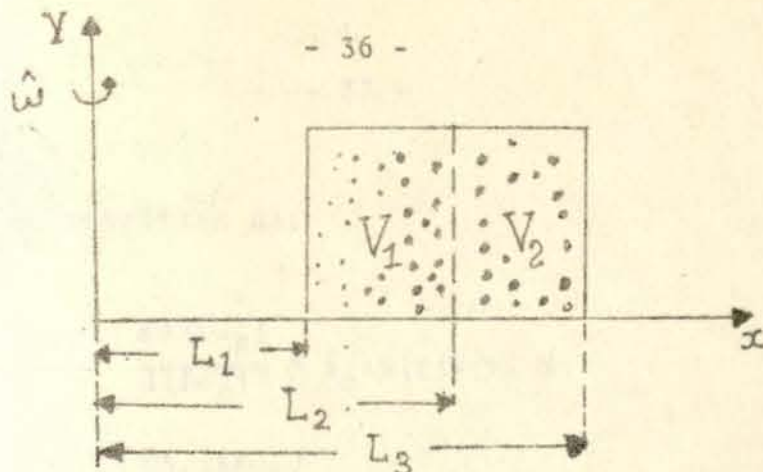


Fig. 5: Centrifugation experimental scheme for perturbing, the system from equilibrium state, in order to establish equations (3.504).

$\hat{\omega}$, rotational velocity; V_1 , volume occupied by particles suspension in distilled water; V_2 , volume occupied by pure water; L_1, L_2 and L_3 are lengths in (cm.) W , partition.

In the above scheme, the volume V_2 is filled by pure water. This cell is placed into centrifugation, the partition is removed and begins to rotate with angular frequency ω_c . After a few moment of centrifugation process, the partition is closed automatically. The process is stopped and each particle in volume V_1 and V_2 will be mixed. So we obtain two different colloidal systems and two different distribution functions $\phi_1(r)$ and $\phi_2(r)$. Obviously each of them depends on the primery function $\phi(r)$ which we have before. These dependences can be written in the form;

$$\left. \begin{aligned} Z_1(\phi(r), \phi_1(r)) &= 0 \\ Z_2(\phi(r), \phi_2(r)) &= 0 \end{aligned} \right\} \dots (3.505)$$

Experimentally it is possible to measure the amplitude of light modulation $A_1(1/\omega_F)$ and $A_2(1/\omega_F)$ for both colloidal systems, and $\phi_1(r)$ and $\phi_2(r)$ can also be calculated. Therefore, equations

(3.504) can be rewritten as;

1. $\frac{dA(1/\omega_F)}{d(1/\omega_F)} = I_0 \Delta K(r) \phi(r) N$
2. $\frac{dA_1(1/\omega_F)}{d(1/\omega_F)} = I_0 \Delta K(r) \phi_1(r) N_1$
3. $\frac{dA_2(1/\omega_F)}{d(1/\omega_F)} = I_0 \Delta K(r) \phi_2(r) N_2 \quad \dots (3.506)$
4. $Z_1(\phi(r), \phi_1(r)) = 0$
5. $Z_2(\phi(r), \phi_2(r)) = 0$
6. $N = N_1 + N_2$

3.6: Equations of Motions

During centrifusion process, the forces that act on the particles are; Inertial and Centripetal forces, $\frac{4}{3} \pi \delta_1 r^3 \omega_c^2 x$ and $\frac{4}{3} \pi \delta_2 r^3 \omega_c^2 x$ respectively. δ_1 and δ_2 are densities of particle and water respectively. The other force, $6\pi r \eta \frac{dx}{dt}$, is the force of friction. Therefore, the equation of motion will be;

$$\frac{dx}{dt} = Cr^2 \omega_c^2 x \quad \dots (3.601)$$

and this gives; $x = x_0 \text{Exp}(Ct^2)$, where ξ is given by;

$$\xi = \begin{cases} \int_0^t \omega_c^2(t) dt; & \text{if } \omega_c = \omega_c(t) \\ \omega_c^2 t & ; \quad \text{if } \omega_c \text{ is constant.} \end{cases}$$

3.7. Deduction of Functional Correlations

The functions $\phi_1(r)$ and $\phi_2(r)$ depends not only on r , but also on $\omega_c(t)$. So they are functions of the parameter ξ . From equation (3.601) follows that, $\phi_1 = \phi_1(r, \xi)$ and $\phi_2 = \phi_2(r, \xi)$. If the total number of particles is N , then $N_{L_2 L_1}$ is the number of particles per unite length in volume V_1 before the centrifuge. If a particle of radius r at a given ξ has coordinate L_2 , then in the starting moment of centrifusion its coordinate was;

$$\bar{X}_0 = L_2 \text{Exp}(-Cr^2 \xi) \quad \dots (3.701)$$

It means only particles of radius r , which had coordinates at the start of centrifusion less than $\bar{X}_0$, could remain in V_1 and particles of radius r , which has coordinates in the starting moment of centrifusion greater than $\bar{X}_0$ would leak to volume V_2 . The total number of particles, $n_1(r, \xi) dr$, whose radius is placed in the interval $(r, r+dr)$, in volume V_1 at a given ξ is;

$$n_1(r, \xi) dr = \begin{cases} \frac{N}{L_2 - L_1} (L_2 e^{-Cr^2 \xi} - L_1) \phi(r) dr; & \text{if } r < \rho \\ 0; & \text{if } r > \rho \end{cases} \quad \dots (3.702)$$

where n_1 is the linear density. ρ is the radius of particles which crossed the distance from L_1 to L_2 at the given ξ . Substituting L_2 and L_1 in equation (3.701), the value of ρ determined is;

$$\rho = \left(\frac{1}{C\xi} \ln \frac{L_2}{L_1} \right)^{1/2} \quad \dots (3.703)$$

The total number of particles $N_1(\xi)$ remaining in the volume V_1 , after centrifusion is;

$$N_1(\xi) = \int_0^{\rho} n_1(r, \xi) dr \quad \dots (3.704)$$

The distribution function in volume V_1 , after centrifusion processes is given by;

$$\phi_1(r, \xi) = \begin{cases} \frac{L_2}{L_1} e^{-cr^2\xi} - 1) \phi(r) \\ \int_0^{\rho} \frac{L_2}{L_1} e^{-cr^2\xi} \phi(r) dr \end{cases} ; \text{ if } r < \rho$$

$$\phi_1(r, \xi) = \begin{cases} 0 \end{cases} ; \text{ if } r \geq \rho \quad \dots (3.705)$$

Equation (3.705) establishes the relation between $\phi(r)$ and $\phi_1(r, \xi)$ and it is one of the relations in equation (3.506).

The number of particles, $n_2(r, \xi)dr$, having radius in the interval $(r, r+dr)$, in volume V_2 is determined from the relation

$$n_2(r, \xi)dr = N\phi(r)dr - n_1(r, \xi)dr \quad \dots (3.706)$$

where $n_1(r, \xi)dr$ is given by equation (3.702).

Rewriting equation (3.706) as;

$$n_2(r, \xi)dr = \begin{cases} \frac{N}{L_2-L_1} (L_2 - L_2 e^{-cr^2\xi}) \phi(r)dr & ; \text{ if } r < \rho \\ 0 & \text{ if } r \geq \rho \end{cases}$$

the total number of particles in volume V_2 at a given ξ is given by;

$$N_2(\xi) = N - \int_0^{\rho} \frac{N}{L_2-L_1} (L_2 e^{-cr^2\xi} - L_1) \phi(r)dr \quad \dots (3.707)$$

The distribution function in volume V_2 is determined to be;

$$\phi_2(r, \xi) = \begin{cases} \frac{\frac{L_2}{L_1}(1 - e^{-cr^2\xi})\phi(r)}{\frac{L_2}{L_1} - 1 - \int_0^{\rho} \left(\frac{L_2}{L_1} e^{-cr^2\xi_1}\right)\phi(r)dr} ; & \text{if } r \geq \rho \\ 0 ; & \text{if } r < \rho \end{cases} \quad \dots (3.708)$$

3.8 Determination of the Distribution Function from Experimental Data Using the Functional Correlations

In order to obtain a working equation for experimental data, some notations are used, these are;

1. $f_1(r, \xi) = N_1(\xi) \Delta K(r) \phi_1(r, \xi)$
2. $f_2(r, \xi) = N_2(\xi) \Delta K(r) \phi_2(r, \xi)$
3. $N_1(\xi) = Nu$
4. $N_2(\xi) = Nt$
5. $\frac{L_2}{L_1} - 1 = a$
6. $\int_0^{\rho} \left[\frac{L_2}{L_1} e^{-cr^2\xi} - 1 \right] \phi(r) dr = Q.$

Referring to equation (3.506); when the first equation is divided by the second and also by the third, the results obtained are;

$$\frac{dA\left(\frac{1}{\omega_F}\right) \cdot \frac{N_1(\xi)}{N}}{dA_1\left(\frac{1}{\omega_F}\right)} = \frac{f(r)}{f_1(r, \xi)} u = \frac{\phi(r)}{\phi_1(r, \xi)} \quad \dots (3.801)$$

and

$$\frac{dA\left(\frac{1}{\omega_F}\right) \cdot \frac{N_1(\xi)}{N}}{dA_2\left(\frac{1}{\omega_F}\right)} = \frac{f(r) t}{f_2(r, \xi)} = \frac{\phi(r)}{\phi_2(r, \xi)} \quad \dots (3.802)$$

Using equations (3.705) and (3.708), Q can be written as;

$$Q = \frac{\left(\frac{L_2}{L_1} e^{-cr^2\xi} - 1\right) f(r) u}{f_1(r, \xi)} \quad \dots (3.803)$$

Because of the relation $N_1 + N_2 = N$, then $u + t = 1$. Substituting $t = 1 - u$, in equation (3.802) and using equation (3.801), the value of u obtained is;

$$u = \frac{a + \left(\frac{L_2}{L_1} e^{-cr^2\xi} - \frac{L_2}{L_1}\right) \frac{f(r)}{f_2(r, \xi)}}{\left(\frac{1}{f_1(r, \xi)} + \frac{1}{f_2(r, \xi)}\right) e^{-cr^2\xi} - \left(\frac{1}{f_1(r, \xi)} + \frac{L_2}{L_1 f_2(r, \xi)}\right)}$$

Combining this last equation with equation (3.801) the result obtained is;

$$\int_0^{\rho} \left(\frac{L_2}{L_1} e^{-cr^2\xi} - 1\right) \phi(r) dr = \psi(\xi) \quad \dots (3.805-a)$$

where,

$$\psi(\xi) = \frac{\left(\frac{L_2}{L_1} e^{-cr^2\xi} - 1\right) \left\{ \frac{L_2}{L_1} - 1 + \frac{L_2}{L_1} (e^{-cr^2\xi} - 1) \right\} \frac{f(r)}{f_2(r, \xi)}}{\left(1 + \frac{f_1(r, \xi)}{f_2(r, \xi)}\right) e^{-cr^2\xi} - \left(1 - \frac{L_2}{L_1} \frac{f_1(r, \xi)}{f_2(r, \xi)}\right)} \quad \dots (3.805-b)$$

In order to determine $\psi(\xi)$; the values of $r = \frac{1}{B\omega_F}$, $L_1, L_2, f(r)$ and $f_1(r, \xi)$ have to be determined. The functions $f(r)$, $f_1(r, \xi)$ and $f_2(r, \xi)$ are the tangents to the curves of equations 3.506-1, 3.506-2 and 3.506-3 respectively. Thus the function $\phi(r)$, we are interested in, is obtained from equation (3.805-a). As the analytical solution

of the integral equation wasn't found, a numerical method was used instead. Rewriting equation (3.805-a) as;

$$\int_0^{\rho} \left[\left(\frac{L_2}{L_1} \right) \frac{1-r^2}{\rho^2} - 1 \right] \psi(r) dr = \psi(\xi) \quad \dots (3.806)$$

The solution of equation (3.806) was obtained by dividing ρ into n equal intervals of length " h ", such that $r_j = j \cdot h$; where $j = 1, 2, \dots, n$. Replacing equation (3.806) by a terminal sum and measuring $\psi(\xi)$ for n - points, the system of equations obtained is;

$$\sum_{k=1}^i a_{ik} \left[\left(\frac{L_2}{L_1} \right) \frac{1 - \left(\frac{k}{i} \right)^2}{\rho^2} - 1 \right] \psi(kh)h = \psi(\xi_i) \quad \dots (3.807)$$

where a_{ik} is the coefficient of applying quadrature formula. Then ξ_i is the value at which $\rho = ih$.

For experimental determination of $\psi(\xi)$, the functions $f_1(r, \xi)$ and $f_2(r, \xi)$ were calculated. In order to reduce the effect of the sticking of particles to the wall of the centrifusion apparatus, $f_2(r, \xi)$ was calculated from the relation $f_1(r, \xi) + f_2(r, \xi) = f(r)$.

3.9, Experimental Results and Discussion

This method was used for determination of the distribution function in size of the practically important specimen, a dispersion of synthetic diamond. The particles, being in water, are characterized by the following parameters;

$$\eta = 10^{-2} \text{ CGS}; \delta_1 = 3.6 \frac{\text{gm}}{\text{cm}^3}; \delta = - \frac{\text{gm}}{\text{cm}^3};$$

$$\mu_1 = 1.9 \times 10^{-3} \text{ CgsE}, \beta = 85^\circ; E = 0.7 \text{ CgsE}$$

$$[P] = 6$$

From equation (3.102) the value of β is found out, by measuring the dipolemoment, μ , by the help of equation (3.101). In this case E , n , and ω are measured experimentally. The value of V is connected with the value of τ , through equation (3.301). The method of determination of τ and α ; is explained in 3.4 and 3.11, from which follows, μ and as a result the value of β can be found out for calculating the value of B from equation (3.106).

The experimental results are given in Tables 2, 3, 4, 5, 6, 7, 8, 9, 10 and 11. The function $\psi(\xi)$ which is found out from experiment is plotted in Fig. 7 and the curve of the distribution function, $\phi(r)$, is plotted in Fig. 8.

The result obtained by electronmicroscop is compared with the result obtained by Electrooptical method. In the former case, 3000 particles were used. Comparing the two methods the results obtained are in good agreement with the physical distribution. Therefore, the method, Electrooptical method of determination of SDF, can be applied to a real distribution.

3.10 CELL DESIGN

The rotating electric field is produced in the cell shown in Fig. 6.

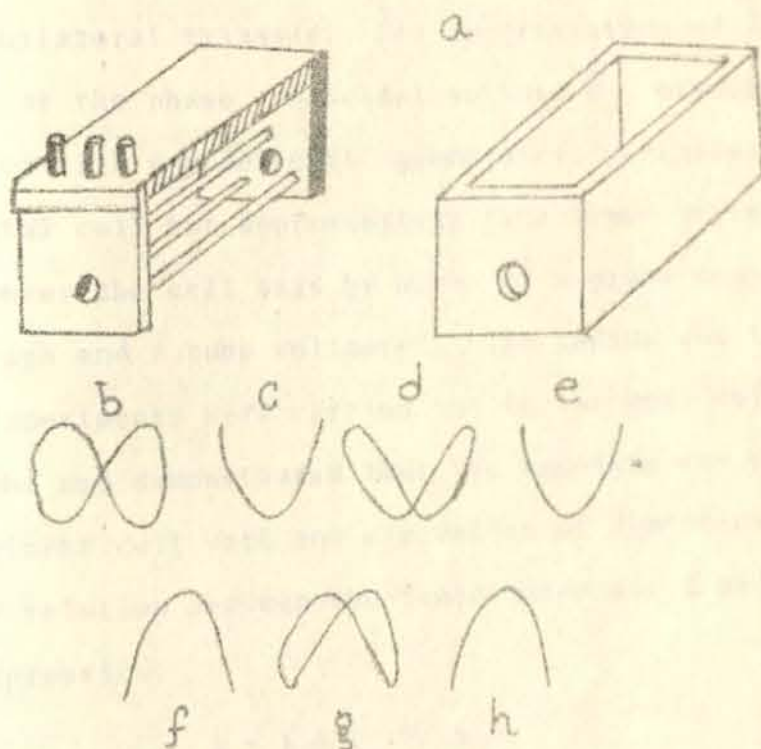


Fig. 6: a) Electrode holder and cell; b, c, d, e, f, g, h, i are Lissajous figures of the second order obtained in the measurement of the lag angle α .

The cell contains a special block with three nickel electrodes mounted in two polytetrafluoroethylene holders. The electrodes are three rods 3mm in diameter and 45-150mm long. The electrode

axes are parallel, and their projections on a perpendicular plane correspond to corners of an equilateral triangle with 10-mm sides. When these electrodes are connected to the phases of a three-phase sinusoidal voltage source with frequency ω , an electric field of constant intensity E , but rotating with angular velocity ω , is produced on the axis passing through the center of the equilateral triangle. For determination of E at a given amplitude of the phase sinusoidal voltage U_0 , experiments were carried out with a model cell, geometrically similar to the experimental cell but approximately five times as large. E was measured near the cell axis by means of a probe connected to an oscillograph and a tube voltmeter. The medium was tap water. Control experiments were carried out in the real cell with a small probe and demonstrated that the modeling was correct. For the experiment cell with the electrodes of dimensions specified above the relation between the field intensity E and U_0 is given by the expression;

$$E = 1.4 U_0 \text{ V/cm.}$$

3.11 Method of Measuring the Lap Angle α

Use of the phases of the three-phase voltage $U = U_0 \sin(\omega t + \phi)$, supplied to the cell, e.g., phase 1 is applied to the horizontal input of the oscillograph. Then the optical sinusoidal signal of frequency 2ω will be scanned as a sinusoid of frequency ω and Lissajous figure of the second order appears

on the oscillograph's screen. The form of this figure will depend on the position of the axis of the polarizer P. The latter is now turned so that the Lissajous figure acquires the form of the characteristic "pit" (Fig. 6c). This means that at the instant when the voltage of phase 1 is passing through zero (the vector $\vec{E}$ in the apparatus is Horizontal) the axis of maximum absorption of the analyzer is parallel to the transmission axis of the polarizer P. (It is assumed that the oscillograph plates are connected so that light causes an upward deflection of the oscillograph beam.) We measured the angle of lag α of the absorption axis of the colloidal particles behind the field E. In practice this angle is measured much more simply and accurately: phases 2 and 3 are changed over by means of a switch in Fig. 1.

Then the field begins to rotate in the opposite direction and the Lissajous figure becomes decomposed (Fig. 6b). P is now turned through an angle θ such that the figure again becomes a pit (Fig. 6e). It is easy to see that the required lag angle is $\alpha = \theta/2$. P should be turned in the direction of the original rotation of the field. If it is found that $\theta > 180^\circ$, this means that the transmission axis and not the absorption axis of the analyzer or of the colloidal particles lags behind the vector of the field E by an angle $\alpha < 90^\circ$. In that case all the procedure described above should be performed with a Lissajous figure having the form of "hump" and not of a pit (Fig. 6f, g and h). The precision in determination of α in a favorable case reaches

$\alpha = 0.5^\circ$. In order to obtain the rotating electric field the oscillator was built. The oscillator consists of three resistance - coupled amplifiers in a ring circuit. Self excitation of this circuit produces sinusoidal oscillations at the anodes, three tubes with a relative phase shift of exactly 120° .

3.1 Procedure

1. The apparatus shown in Fig. 1, was constructed.
2. The sample in distilled water was illuminated using a white light source (ADZ - 50 DC lamp).
3. Using a three-phase voltage source, a rotating field E of magnitude 0.7 CGS was applied.
4. A modulated intensity was observed on the screen of the oscillograph "O₁". After amplification by the photomultiplier A, the modulated intensity was measured (Relative units).
5. Keeping E at constant value, the rotational frequency was increased and for each increment the amplitude was measured (Table 2).
6. The values of f and the corresponding sizes were calculated using equation (3-213). Relative units are used for the values of " f ".
7. The system is perturbed using centrifusion experiment Fig. 5 and
 - a) The cell with the sample was set into a

rotation (without the partition) at $\omega_c = 314.2$
(sec)⁻¹ and for each interval of $t = 6$ min. Step
5 was repeated for the particles in volume V_2 upto
 $t = 54$ minutes.

- b) The function f_2 and the corresponding sizes were
calculated using the relations;

$$\frac{dA_2(1/\omega_F)}{d(1/\omega_F)} = I_0 f(1_{cr})$$

and equations (3-805); relative units are used and
the results are given in Tables 3,4,5,6,7,8,9 and 10.

8. The distribution function was determined using equation
(3-807) and the tables above by simpsons method of
computation (Table 11).

TABLE 2

$t = \infty, \xi = \infty$

No.	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
$10^{-2} \omega_{-1}$ (sec)	9.42	4.71	3.14	2.32	1.88	1.57	1.34	1.18	1.04	1.00	0.84	0.76	0.72	0.67	0.63
$10^3/\omega$ sec	1.1	2.1	3.2	4.3	5.3	6.4	7.44	8.44	9.60	10.47	11.90	12.73	13.84	15.01	15.92
$r \times 10^5$ cm.	3.1	6.3	9.4	12.4	15.6	18.8	22.9	25.0	28.3	30.9	35.3	37.6	40.0	44.4	47.0
A rel-un	13	25	42	48	53	58	63	66	69	72	76	80	80	80	79
f rel-un	55	46	28	22	18	17	13	10	9	7	5	2	0	0	0

TABLE 3

$$t = 6\text{min}; \xi = 3.54 \times 10^7 \text{ sec}^{-1}, \omega_c = 314.2 \text{ sec}^{-1}$$

No.	1	2	3	4	5	6	7	8	9	10	11
$10^{-2} \omega$ (sec) ⁻¹	5.15	2.89	2.07	1.62	1.32	1.11	0.96	0.85	0.76	0.69	0.63
$10^3 \omega$ sec	1.94	3.46	4.83	6.19	7.58	9.00	10.40	11.80	13.15	14.60	15.91
$r \cdot 10^5$ cm.	5.70	10.20	15.50	18.90	22.40	26.60	30.80	34.90	38.90	43.10	47.00
A rel-un	10.00	21	30	34	38	40	42	43	45	44	45
f_2 rel-un	32	29	20	16	15	11	7	3	1	0	0

TABLE 4

$t = 12\text{min}$, $\epsilon = 3.54 \times 10^7 \text{ sec}^{-1}$, $\omega_c = 3.14.2 \text{ sec}^{-1}$

No.	1	2	3	4	5	6	7	8	9	10	11
$10^{-2} \omega_{-1}$ (sec)	6.47	3.20	2.20	1.68	1.35	1.13	0.98	0.85	0.76	0.69	0.63
$10^3/\omega$ sec.	1.55	3.12	4.55	5.96	7.40	8.84	10.20	11.70	13.15	14.60	15.91
$r \times 10^5$ cm.	9.6	9.2	13.4	17.6	21.8	26.2	30.2	34.6	38.9	43.1	47.0
A rel-un	12	23	27	35	40	44	47	48	49	49	50
f_2 rel-un	35	24	14	11	9	7	6	3	2	0	0

TABLE 5

$$t = 18\text{min}, \xi = 1.06 \times 10^8 \text{ sec}^{-1},$$

No.	1	2	3	4	5	6	7	8	9	10	11
$10^{-2} \omega$ (sec) ⁻¹	6.94	3.89	2.31	1.64	1.26	1.04	0.84	0.76	0.67	0.60	0.55
$10^3 / \omega$ sec	1.54	2.57	4.32	6.10	7.96	9.65	12.00	13.15	14.87	16.75	18.29
10^5xr cm.	4.6	7.5	12.8	18.0	20.0	28.5	35.2	38.9	44.0	49.4	53.9
A_2 rel-un	19	30	34	40	47	51	53	54	54	55	56
f_2 rel-un	41	35	32	29	11	7	3	2	0	0	0

TABLE 6

$$\tau = 30\text{min}, \xi = 1.78 \times 10^8 \text{ sec.}^{-1}$$

No.	1	2	3	4	5	6	7	8	9	10	11
$10^{-2} \omega$ (sec) ⁻¹	9.42	2.01	2.87	2.00	1.51	1.26	1.03	0.89	0.78	0.69	0.63
$10^3 / \omega$ sec	1.06	4.97	3.49	5.00	6.6	7.96	9.70	11.29	12.84	14.47	15.92
$r \times 10^5$ cm.	3.1	5.7	25	31	40	49	52	53	53	54	55
A_2 rel-un	8	15	25	31	40	49	52	53	53	54	55
f_2 rel-un	43	39	24	20	15	11	7	4	2	0	0

TABLE 6

$$t = 30 \text{ min}, \xi = 1.78 \times 10^8 \text{ sec.}^{-1}$$

No.	1	2	3	4	5	6	7	8	9	10	11
$10^{-2} \omega$ (sec) ⁻¹	9.42	2.01	2.87	2.00	1.51	1.26	1.03	0.89	0.78	0.69	0.63
$10^3/\omega$ sec	1.06	4.97	3.49	5.00	6.6	7.96	9.70	11.29	12.84	14.47	15.92
$r \times 10^5$ cm.	3.1	5.7	25	31	40	49	52	53	53	54	55
A_2 rel-un	8	15	25	31	40	49	52	53	53	54	55
f_2 rel-un	43	39	24	20	15	11	7	4	2	0	0

TABLE 7

$$t = 36\text{min}, \xi = 2.12 \times 10^8 \text{ sec}^{-1}$$

No.	1	2	3	4	5	6	7	8	9	10	11
$10^{-2} \omega$ (sec) ⁻¹	9.42	5.67	2.98	2.05	1.54	1.23	1.00	0.89	0.77	0.70	0.63
$10^3/\omega$ sec	1.06	1.76	3.35	4.87	6.50	8.16	9.65	11.30	13.05	14.21	15.92
$r \times 10^5$ cm.	3.1	5.2	10.0	14.4	19.3	24.2	28.5	33.5	38.5	39.5	47.0
A_2 rel-un	9	15	25	33	42	50	51	55	54	55	56
f_2 rel-un	44	40	25	10	15	11	8	4	2	0	0

TABLE 8

$$t = 42\text{min}, \epsilon = 2.12 \times 10^8 \text{ sec}^{-1}$$

No.	1	2	3	4	5	6	7	8	9	10	11
$10^{-2} \omega$ (sec) ⁻¹	9.42	6.16	3.20	2.09	1.56	1.25	1.04	4.89	0.79	0.70	0.63
$10^3/\omega$ Sec	1.06	1.62	3.12	4.78	6.40	8.00	9.89	11.21	12.73	14.34	15.92
$r \times 10^5$ cm	3.1	4.8	9.2	14.5	19.0	24.0	28.2	32.4	37.7	42.9	47.0
A_2 rel-un	10	19	29	30	44	52	53	54	57	58	58
f_2 rel-un	48	41	24	21	15	12	7	3	1	0	0

TABLE 9

$$t = 48\text{min}, \xi = 2.84 \times 10^8 \text{ sec}^{-1}$$

No	1	2	3	4	5	6	7	8	9	10	11
$10^{-2} \omega$ (Sec) ⁻¹	9.42	6.47	3.15	2.11	1.55	1.26	1.05	0.90	0.79	0.70	0.63
$10^3 / \omega$ Sec.	1.06	1.55	3.18	4.74	6.44	7.96	9.53	11.13	12.73	14.34	15.92
$r \times 10^5$ cm.	3.1	4.6	9.2	14.0	19.2	23.5	28.2	32.9	37.6	42.7	47.0
A_2 rel-un	11	22	31	39	44	48	49	55	59	59	60
f_2 rel-un	48	46	24	21	16	11	9	4	1	0	0

TABLE 10

$$t = 54\text{min}, \xi = 3.19 \times 10^8 \text{ sec}^{-1}$$

No.	1	2	3	4	5	6	7	8	9	10	11
$10^{-2} \omega$ (Sec) ⁻¹	9.42	6.79	3.24	2.13	1.6	1.31	1.09	0.92	0.80	0.70	0.63
$10^3 / \omega$ Sec.	1.06	1.47	3.1	4.69	6.24	7.65	9.15	10.90	12.53	14.21	15.92
$r \times 10^5$ cm.	3.1	4.4	9.1	13.8	18.6	22.8	27.0	32.2	37.0	42.0	47.0
A_2 rel-un	8	16	27	37	44	45	51	58	59	62	63
f_2 rel-un	51	48	25	21	14	12	8	6	2	0	0

TABLE 11

$\xi \times 10^{-7}$ (Sec) ⁻¹	$\Psi(\xi)$	$\frac{\Delta\Psi}{\Psi}$	$r \times 10^5$ cm	$\phi(r) \cdot 10^{-4}$ cm ⁻¹	$\frac{\Delta\phi}{\phi}$
0	.18	-	0	0	-
3.54	.25	.05	1.0	.38	.05
7.09	.15	.04	2.0	1.27	.07
10.6	.11	.06	3.0	1.52	.08
17.8	.07	.08	4.0	1.65	.08
21.2	.04	.08	6.0	1.28	.08
24.8	.03	.08	8.0	.75	.08
28.4	.025	.08	10.0	.53	.09
31.9	.02	.08	12.0	.35	.10

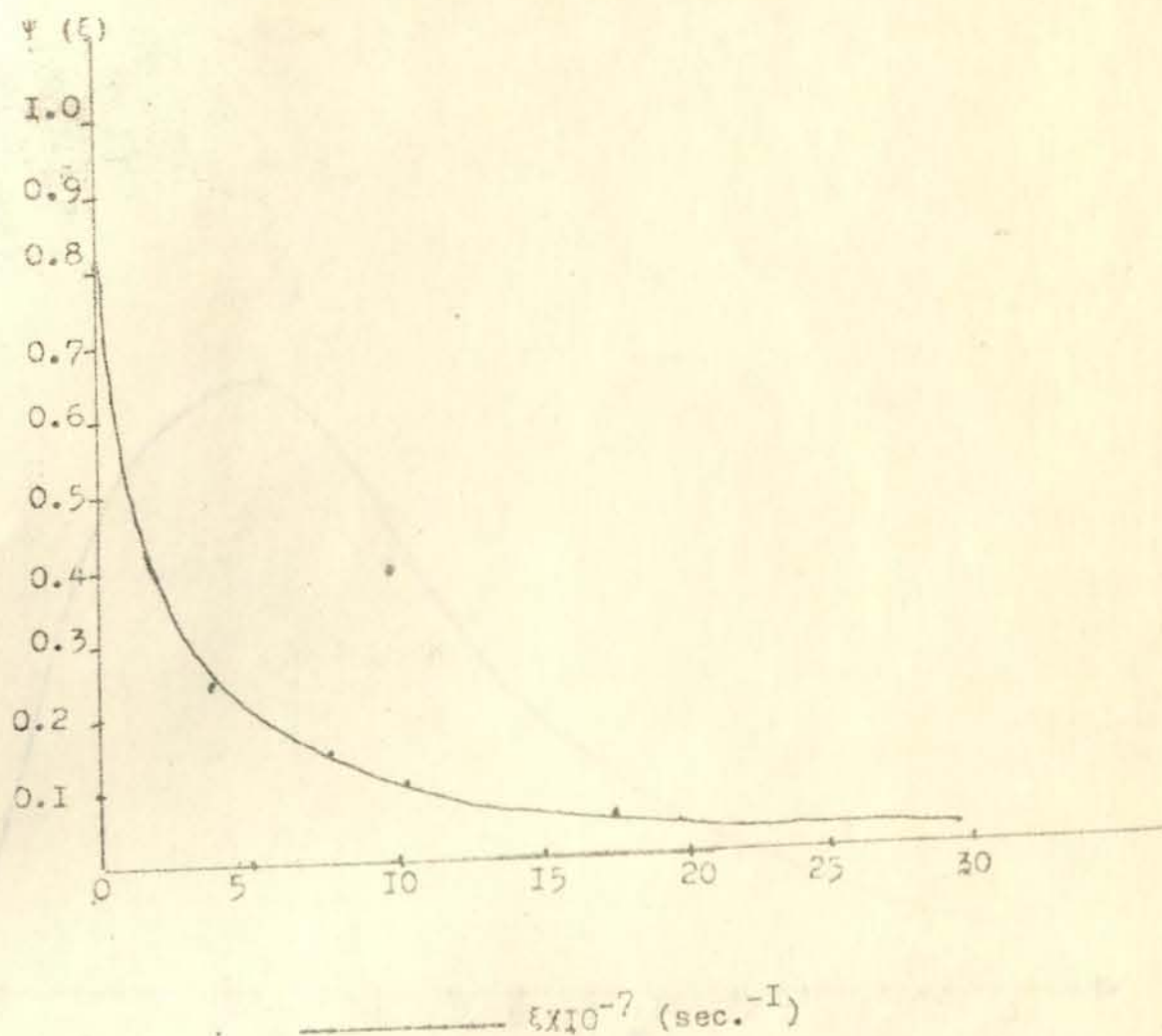


Fig. 7

The curve of $\Psi(\xi)$ versus the centrifusion parameter ξ .

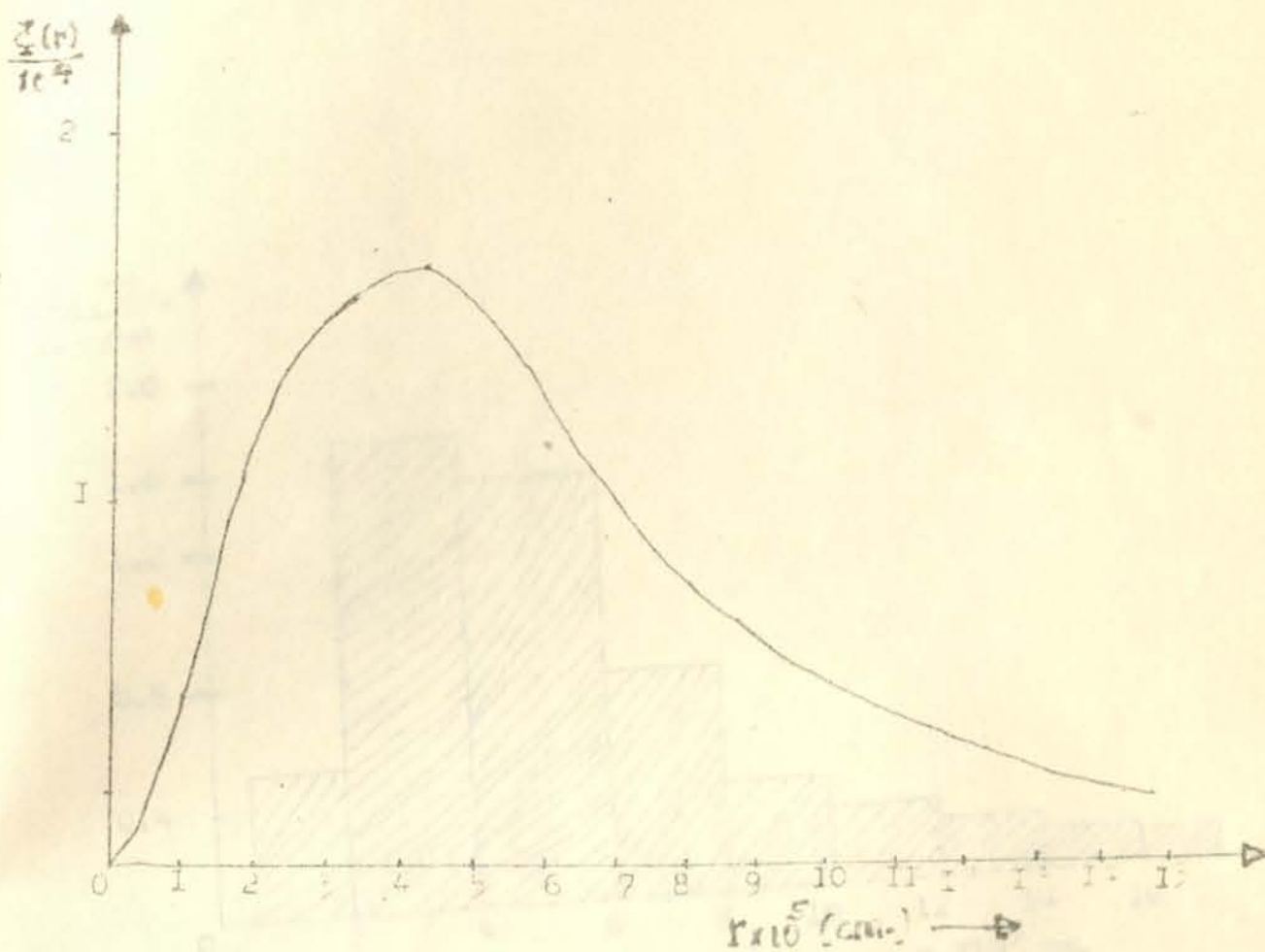


Fig. 8: Plot of the Distribution Function in size.

Fig. 9: Size distribution function

determined by Electromicroscopy
for 5000 particles.

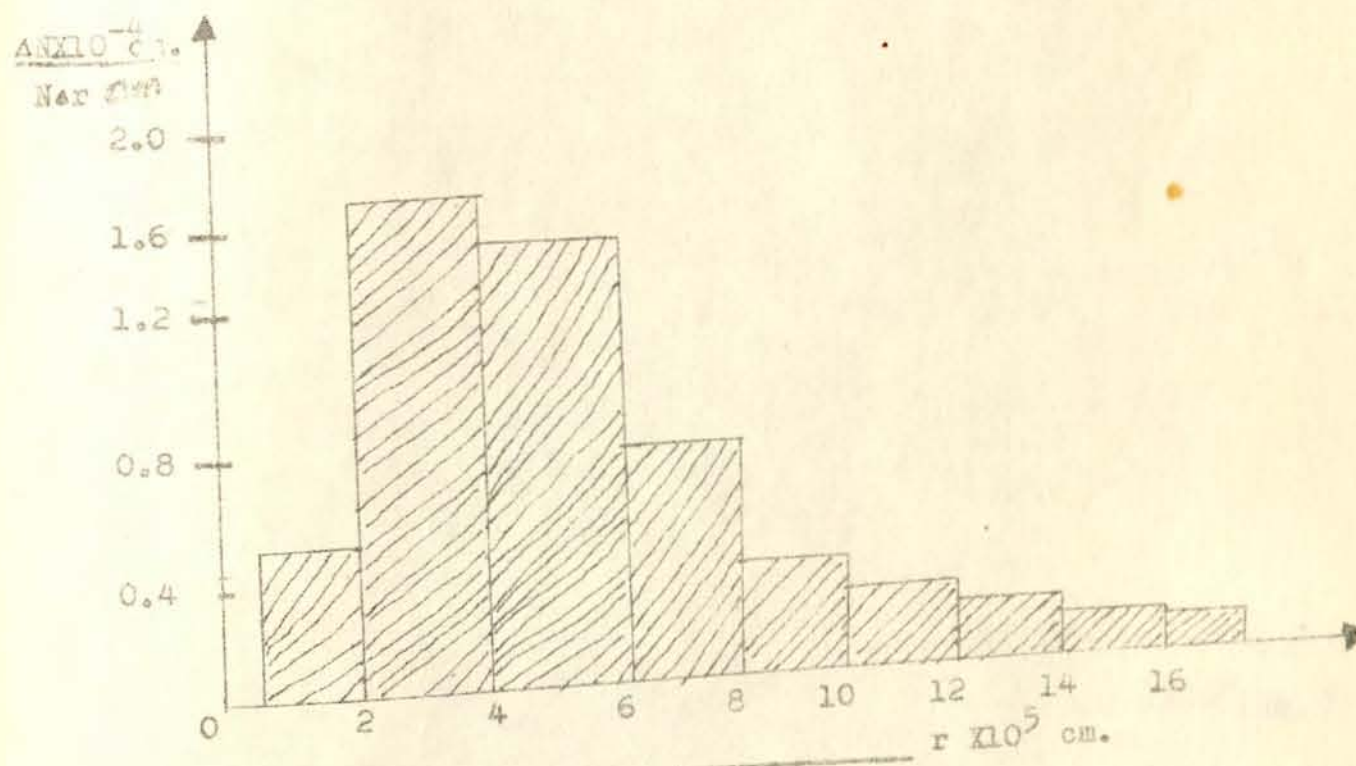


Fig. 9 : Size distribution function
determined by Electronemicroscope
for 3000, particles.

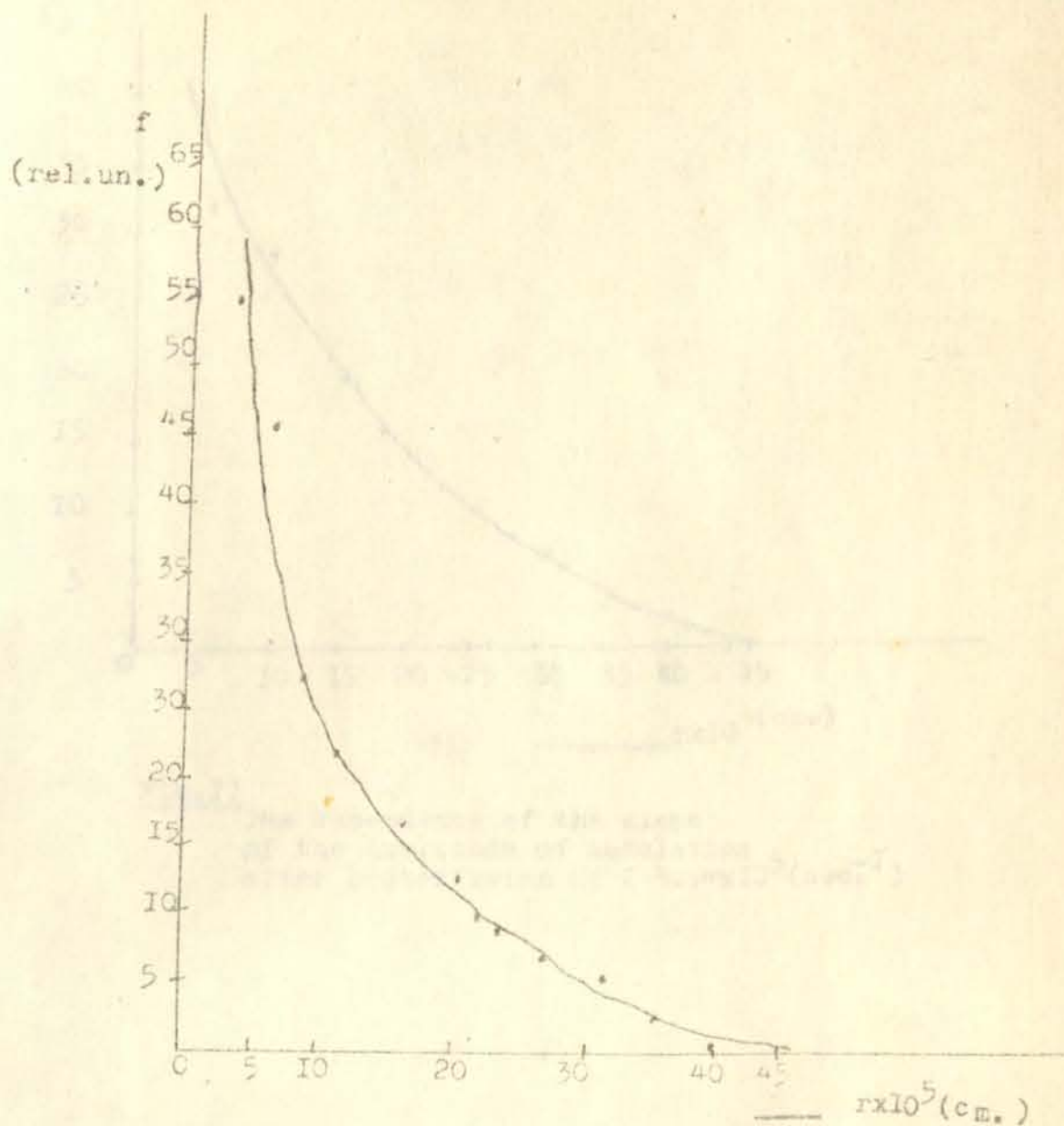


Fig. 10

The graph of the slope of
the Amplitude of Modulation
for the unperturbed system.

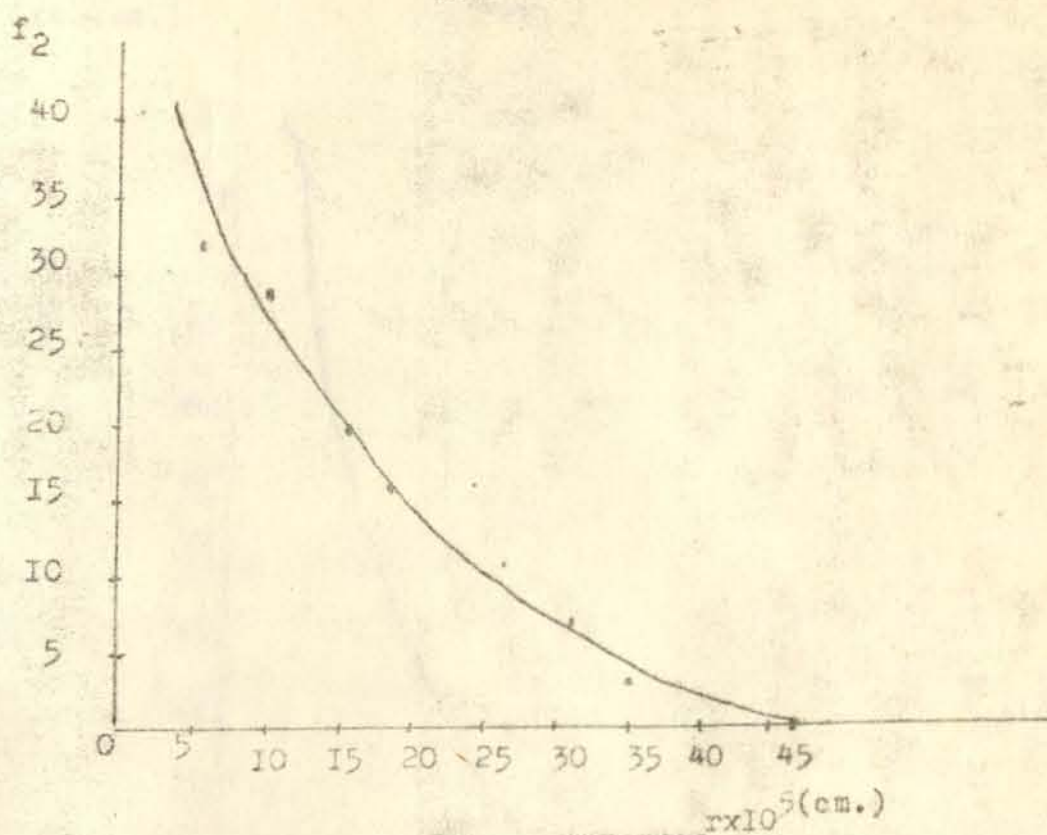


Fig. II

The dependence of the slope
of the amplitude of modulation
after centrifugation at $\omega = 3.54 \times 10^5 \text{ (sec.}^{-1}\text{)}$

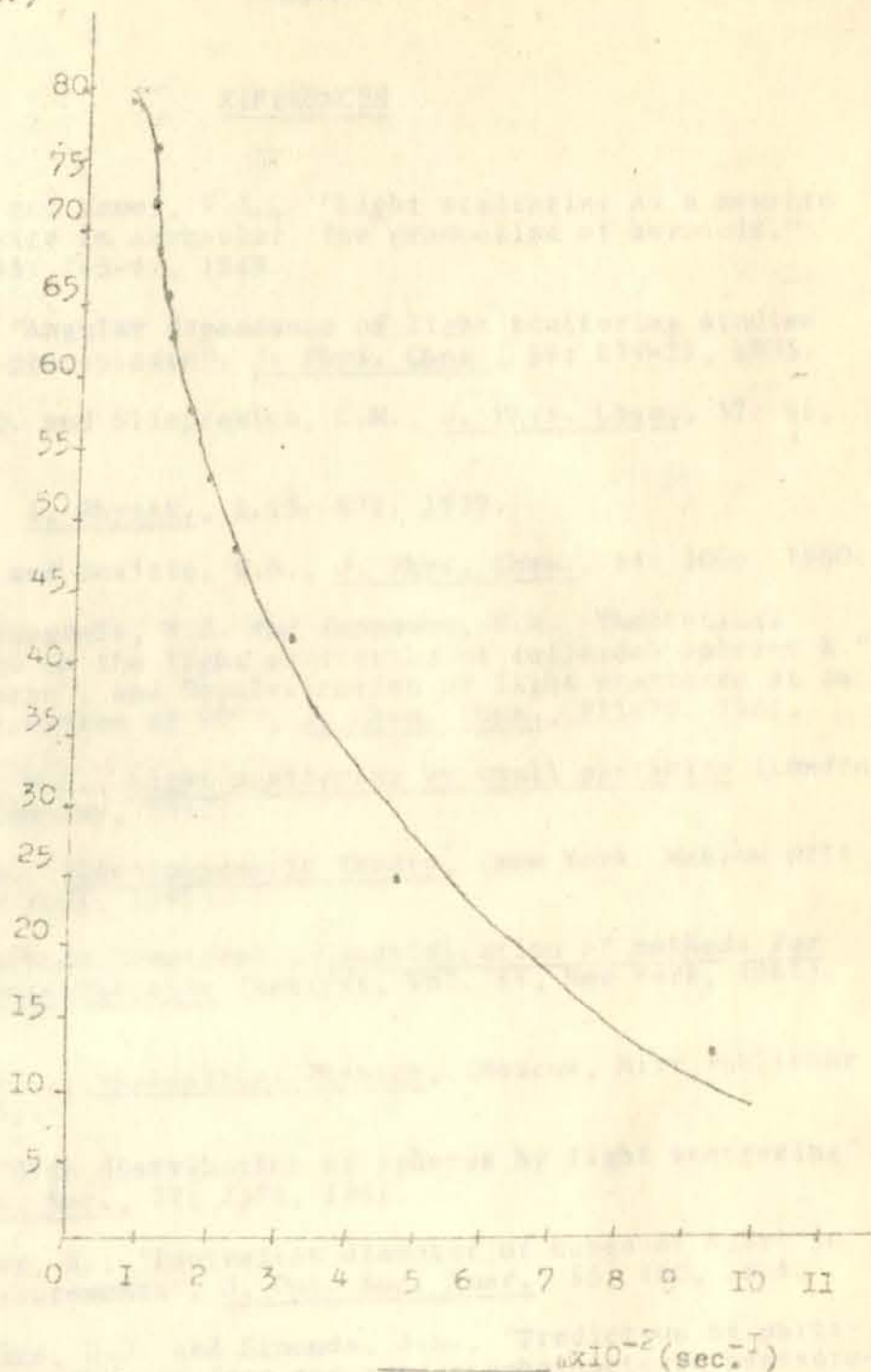


Fig. 12

The Amplitude of modulation curve in dependence on the field frequency; For the equilibrium state of the system

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DECLARATION

I, the undersigned declare that the thesis is my original work and has not been presented for a degree in any other University. All sources of materials used for the thesis have been duly acknowledged.

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