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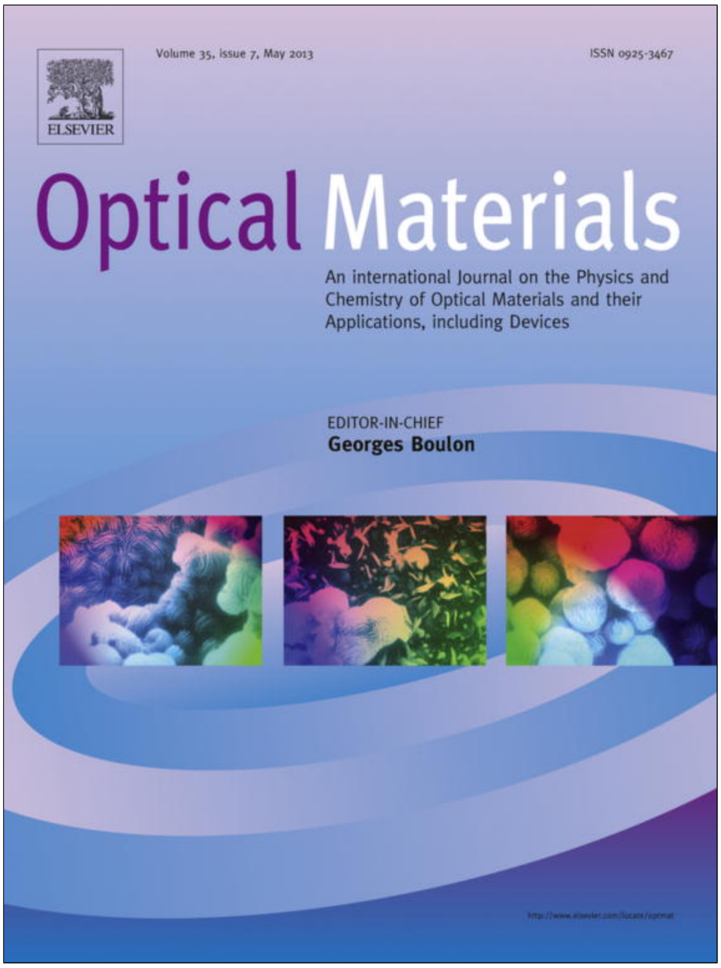
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Polarization dependent extinction coefficients of superparamagnetic colloids in transverse and longitudinal configurations of magnetic field

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ABSTRACT

We review here, our work on the light scattering by ferrofluids and mixture of ferrofluids and other non-magnetic suspensions subjected to an external static magnetic field. Detailed derivation of extinction coefficients of the systems when incident light is propagating along transverse and longitudinal direction of the field are carried out. In case of inclusion of anisotropic diamagnetic micron sized particles in a ferrofluids, effects of'magnetic holes' generated in the system on the extinction are discussed. The present work is analyzed in light of other similar investigations. It is shown that the study of polarization dependence of extinction coefficients in transverse field configuration is useful to arrive at an unambiguous conclusion regarding chain formation in a ferrofluid.

1. Introduction

Ferrofluids are complex fluids. Their complexity arises when they are subjected to static, gradient or rotating magnetic fields. Single domain nanomagnetic particles stably dispersed in a carrier liquid play a pivotal role in ascribing interesting as well as potentially useful properties to the fluids. Magnetization in such a particle is saturated hence; it behaves like a tiny magnet. A magnet naturally follows the direction of applied magnetic field. Consequently, a large number of tiny magnets in a ferrofluids transmit drag to surrounding liquid in a gradient field and generate ferrohydrodynamic body force. This force is responsible for several novel phenomena in hydrostatics and hydrodynamics of ferrofluids and has also made possible an extensive number of engineering and technological applications [1–4]. This growth has almost superseded interest in research on properties and applications in uniform and rotating magnetic fields. Recently work on magneto-viscous effects in rotating field and optical effects in static field has aroused interest in other nonconventional applications [5–8]. In this paper, we shall describe magnetically induced optical effects in ferrofluids as well as dispersions of microscopic particles in ferrofluids.

Magneto-optical effects in magnetic colloids were studied much earlier before the advent of ferrofluids. Majorana was the first to discover magneto-birefringence and dichroism in an aged iron oxide colloid called 'Bravis iron' [9]. He also found that (i) the effects are proportional to square of the applied magnetic field and (ii) in a birefringent dichroic colloid; the linearly polarized wave which was retarded more was also absorbed to a greater extent ('Majorana Rule'). The effects were attributed to the suspended particles by Schmauss [10] and to the ultramicroscopic nature of the particles by Cotton and Mouton [11]. They also observed the relation $(n_e - n) = 2(n - n_o)$ between the two principal refractive indices for extraordinary (n_e) and ordinary (n_o) waves and n stands for the refractive index in zero field. Langevin gave theory of these effects on line of his theory of paramagnetism [12]. Similar relation also found to be applicable for other parameters like turbidity, absorption, etc. [13,14]. Bitter showed that colloidal solution of magnetite can be used to study domain structure on ferromagnetic surfaces [15]. This work has attracted attention of several other workers [16–19]. Their investigations have revealed several interesting optical properties of ferromagnetic suspensions subjected to a static magnetic field. For example, even though individual particles in the suspensions were nanosized that is much smaller than the wavelength of incident light, still Rayleigh scattering theory could not explain the observed magneto-optical transmission in a freshly prepared colloid of magnetite particles. When the same colloid was studied after some aging, it exhibited effects that can be accounted by Rayleigh theory [19]. It was inferred that in the

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Nomenclature

A_j	elements of the scattering matrix of ferrofluid
$\hat{A}$	projected area of large particle
C_{ext}	extinction coefficient
d	diameter of nanomagnetic particle
E	amplitude of electric vector of light
$f(\theta, \varphi)$	orientation distribution function
H	magnitude of magnetic field in Tesla
h ; h'	dimensionless field parameter for nanomagnetic and diamagnetic particles respectively
k	wave vector
t	path length of light in the sample cell
k_B	Boltzmann constant
l,r	refers to the parameter parallel and perpendicular to the field respectively
κ	subscript 'K' refers to the extinction parameter in longitudinal configuration
$L(h)$	Langevin function
'l';'r'	subscript 'l'; and 'r' refers to the amplitude parallel and perpendicular to the scattering plane
M	magnetic moment of a ferrofluid
m	domain magnetization of a particle
N	number of nanomagnetic particles per millilitre
N'	number of large diamagnetic particles per millilitre
n_p	number of 'p' types of particles per millilitre
n	refractive index of ferrofluid in zero field
n_e, n_o	refractive index of extraordinary and ordinary waves in a ferrofluid
m_j	refractive index of anisotropic particles
j	subscript j (j = 1,2,3) refers to the three principal axes of the particle
P	dielectric polarization of the particle
T	temperature of the medium
τ	subscript τ refers to transverse configuration
U	potential energy of particles in a magnetic field
V	volume of the particle
α	polarizability tensor
θ	polar angle with respect to the direction of propagation 'OZ'
ϕ	azimuth angle with respect to a selected direction in a plane perpendicular to 'OZ'
χ	diamagnetic susceptibility
$\varepsilon(h)_\tau$	combined orientation function for a mixture of diamagnetic particles and a ferrofluid subjected to a transverse magnetic field
Q_τ	ratio of extinction coefficients in field and zero field respectively for ($\tau = L, R$) electric vector in parallel and perpendicular to the field in transverse configuration and $\tau = K$ in longitudinal configuration

freshly prepared colloids the magnetite particles exist in an aggregated state, presumably linked be magnetic attractions between the particles. On subsequent ageing the aggregates dissociate into independent particles. Soon after the preparation of magnetic fluid for its possible use in rocket fuels and subsequent development of science and technology of magnetic fluids which are also called ferrofluids, research interest in magneto optical effects in ferrofluids gained momentum [20–30]. Earlier work on magneto optical effects on magnetic colloids were mainly confined to aqueous colloids and stability of colloids were attained mainly by electrostatic charge. In case of ferrofluids host liquid can be water, hydrocarbons, oils or any synthetic organic liquid like esters, diesters, etc. For non-polar liquids, surfactant coating is used to provide steric repulsion to counteract the van der Walls and magnetic attractions. Moreover, nanomagnets may be any ferrite like magnetite, cobalt ferrite or mixed ferrite. Consequently, magnetically induced effects are more complicated and interpretation of experimental data will depend on the type of ferrofluids used in the investigations. For example, when ferrofluid has a polar base like water, it is more amenable for chain formation while if a non-polar like kerosene is used, chaining can be prevented by appropriate surfactant coating on nanomagnetic particles. Hence, magneto optical effects are quite different in the two cases [31–33]. This fact is sometimes overlooked [34]. It may be remarked here that, in well stabilized water based ferrofluids aggregation can be prevented [35]. It is also reported that magnetically induced birefringence can only be observed if either chains are formed under the influence of the field or they may be pre-existed in the fluid. [36]. In the following sections we shall show that even in absence of chain formation or aggregation the birefringence can be observed provided the particles are anisotropic. In the next Section first we shall describe methods of preparation of ferrofluids and other nonmagnetic suspensions. Experimental assembly useful for the present study will also be described. Subsequently, theory of magneto-optical effects in a ferrofluid as well as nonmagnetic particles suspended in ferrofluid will be developed. Such a binary fluid is also known as inverse ferrofluids. Results will be analyzed on the basis of this theory.

2. Experimental

2.1. Ferrofluids

There is several methods for synthesis of ferrofluids like, co precipitation, sol-jell technique, micro emulsion, hydrothermal, etc. Amongst this co precipitation technique is found to be convenient. Mixtures of stoichiometric solutions of divalent and trivalent metal ions (Fe, Mn, Zn as per requirement) are reduced by alkali solution (NaOH or NH_4OH) under vigorous stirring and the black magnetic precipitates were washed several times with warm distilled water. The particles then coated with a single layer of an appropriate surfactant (oleic acid, lauric acid). These coated particles are dispersed in a liquid carrier like water or kerosene. The fluid is then centrifuged at around 12,000 rpm to remove aggregates. Crystalline nature of the particles is ascertained by X-ray diffraction and saturation magnetization of the fluid is measured with a vibrating sample magnetometer. The average particle size in a typical ferrofluid is about 10 nm. The fluid is then diluted so as to make it sufficiently transparent for optical investigations. Details are described in earlier papers [37,38]

2.2. Suspensions of nonmagnetic particles

Commercially available powder of nonmagnetic materials like graphite is first cleaned with dilute nitric acid to remove impurities and washed with acetone and water. The dried powder is then pulverized in a ball mill in presence of a liquid carrier like kerosene and surfactant like oleic acid. Using fractional sedimentation, suspensions having average size of 3, 2, 1 and 0.5 μm were obtained. The particles were found to be almost spherical [38].

2.3. Experimental assembly

Several set ups are described to measure various magneto-optical parameters of ferrofluids [25,39,40]. A monochromatic collimated light beam is obtained either from a monochromator, laser or an interference filter. The beam is passed through a polarizer and then incident on a sample cell containing the colloid under investigation. The cell is placed between the pole pieces of an electromagnet. For measurements in the longitudinal configuration the pole pieces are bored through. The transmitted light is either passed through a second polarizer or is directly received by a photo detector. A wave plate is sometimes introduced between the two polarisers for measurement of birefringence. Output of the detector is connected either to a lock-in – amplifier or a storage oscilloscope.

3. Optics of ferrofluids

3.1. Scattering by anisotropic dipoles

Consider a diluted ferrofluids in which each nanomagnetic particles are well dispersed. Light propagation through such a fluid will be governed by scattering properties of individual particles. In a diluted fluid, inter-particle distance is large. Hence, single scattering theory will be applicable. Scattering by such a system is described by the scattering matrix $S(\theta, \phi)$ (Refer to Ref. [41] for notations). As a consequence of fundamental extinction formula, light transmitted through the system is determined by scattering matrix of individual particles for the forward direction $\theta = 0$ [41]. Under the action of applied magnetic field, each nanomagnet will be oriented along the direction of applied field and the system will exhibit uniaxial anisotropy with its optic axis along the direction of the field. Consequently, non diagonal terms of the matrix $S(0)$ will be zero and the fluid will exhibit linear anisotropy. Elements $S_1(0)$ and $S_2(0)$ can be computed from anisotropic dipole scattering theory [41]. If an arbitrary scatterer is illuminated by a plane electromagnetic wave propagating along 'OZ' direction (perpendicular to the page), then the amplitudes of electric field of the scattered wave are given by

$$\begin{pmatrix} E_l \\ E_r \end{pmatrix} = \begin{pmatrix} S_2(\theta,\phi) & S_4(\theta,\phi) \\ S_1(\theta,\phi) & S_3(\theta,\phi) \end{pmatrix} \frac{\exp(-ikr+ikz)}{ikr} \begin{pmatrix} E_{lo} \\ E_{r0} \end{pmatrix} \quad (1)$$

In the above equation θ is the polar angle with the direction 'OZ'; ϕ is the azimuth angle with respect to a selected direction in a plane perpendicular to the 'OZ'. The direction of incident wave and the direction $S(\theta, \phi)$ define a scattering plane. E_{l0} and E_{r0} are the components of the electric vector of the incident light, respectively parallel and perpendicular to the scattering plane. The sense of the unit vectors 'r' and 'l' are so chosen that $(r \times l)$ is the direction of propagation. $S_j(\theta, \phi)$ defines the elements of the scattering matrix S which should be determined from the size, shape and electro-magnetic constitution of the nanomagnets. In the case of transmitted light, angles $\theta = 0°$ and $\phi = 0°$.

In a colloidal solution a large number of particles are oriented in random directions. The amplitudes of the electric field components of the emergent light from the colloid will be obtained by adding elements of $S(0)$ matrix of the individual particles. If 'OX' and 'OY' are any two mutually perpendicular directions in a plane perpendicular to 'OZ' then the components E_x and E_y of the transmitted light will be given by

$$\begin{pmatrix} E_x \\ E_y \end{pmatrix} = \begin{pmatrix} 1-A_2 & A_4 \\ =A_3 & 1-A_1 \end{pmatrix} \begin{pmatrix} E_{x0} \\ E_{y0} \end{pmatrix} \quad (2)$$

where $A_j = 2\pi l k^{-2} \sum S_j(0) n_p$, $j = 1,2,3,4$

The summation 'p' is extended for all types of particles and n_p is the number of particles of type 'p' per ml in the colloid, l is the length of light path through the colloid and k is $2\pi/\lambda$ wave number in the medium surrounding the particles.

The matrix A indicates a combination of the effects of (i) linear birefringence, (ii) linear dichroism, (iii) circular birefringence and (iv) circular dichroism. All these effects may not be observed if the matrix A possesses some form of symmetry.

3.2. Cloud of axially symmetric particles

When a ferrofluid is subjected to an external magnetic field nanomagnets are oriented along the direction of the field. The direction of the field and the direction of propagation can be taken as the plane of reference. Mirror image of the particle in this plane is identical to the particle itself. It may be noted that in previous case scattering plane could not be uniquely defined. Scattering matrix of the mirror image is of the form $\begin{pmatrix} S_2 & S_4 \\ -S_3 & S_1 \end{pmatrix}$. Consequently the resultant matrix $(\sum A)$ becomes diagonal. In this case, amplitudes of the electric vector of the transmitted light are given by

$$\begin{pmatrix} E_l \\ E_r \end{pmatrix} = \begin{pmatrix} 1-A_2 & 0 \\ 0 & 1-A_1 \end{pmatrix} \begin{pmatrix} E_{l0} \\ E_{r0} \end{pmatrix} \quad (3)$$

The subscript 'l' and 'r' now refers to the direction parallel and perpendicular to the applied field. It is observed that the transmitted light shows only linear optical anisotropy. The matrix contains four parameters, two real and two imaginary and the extinction coefficients and birefringence are given by

$$(C_{ext})_l = (2/t)\mathrm{Re}A_2 \quad (4)$$

$$(C_{ext})_r = (2/t)\mathrm{Re}A_1 \quad (5)$$

$$(n_l - n_r) = (n_m/kt)(\mathrm{Im}A_2 - \mathrm{Im}A_1) \quad (6)$$

Here,'t' is the length of the light path in the sample cell, n_m is the refractive index of the medium and k is the wave vector. If, the direction of propagation of light coincides with the direction of the applied field then the matrix assumes the form $\begin{pmatrix} 1-A_1 & 0 \\ 0 & 1-A_1 \end{pmatrix}$. The propagation will be scalar, that is the extinction and refractive index will be independent of polarization of light and there will be no birefringence and dichroism and

$$(C_{ext})_K = (C_{ext})_r \quad (7)$$

As stated above nanomagnets have their size $\ll \lambda/2\pi$ and the product of modulus of complex refractive index $|m|$ of the nanomagnet and size is also $\ll \lambda/2\pi$, consequently, scattering by the ferrofluids will be governed by Rayleigh scattering. In this case a nanomagnet is considered as situated in a uniform electric field. Scattered wave is described by the dipole scattering theory. The oscillating electric field of the incident light induces oscillating dipole moment P in the particle given by

$$P = \alpha E \quad (8)$$

The polarizability tensor 'α' is a complex symmetric tensor and it can be determined from geometric shape and electromagnetic constitution of the particle. In the present case the principal axes of the dielectric tensor $\epsilon_j (\epsilon_j = m_j^2)$ coincide with the geometric axes of the nanomagnets and hence, the principal axes of α also coincide with the geometric axes and the principal components of α are given by

$$\alpha_j = (V/4\pi)\left(\frac{1}{L_j + \left(\frac{1}{\varepsilon_j - 1}\right)}\right) \quad (9)$$

Here, L_j are the three depolarization factors. Since, particles are assumed to be uniaxial $\alpha_2 = \alpha_3 = \alpha_o$ and $\alpha_1 = \alpha_e$.

Suffix 'e' and 'o' represent extraordinary and ordinary axes respectively. It may be remarked here that a nanomagnet resembles an oblate spheroid and hence $\alpha_e > \alpha_o$.The scattering matrix S0 of a ferrofluid will be given by

$$\begin{pmatrix} 1-A_2 & 0 \\ 0 & 1-A_1 \end{pmatrix} = ik^3 \begin{pmatrix} P_2 & 0 \\ 0 & P_1 \end{pmatrix} \quad (10)$$

$$P_1 = C_{11}^2 \alpha_o + (C_{12}^2 + C_{13}^2)\alpha_e$$

$$P_2 = C_{21}^2 \alpha_o + (C_{22}^2 + C_{23}^2)\alpha_e$$

Here, C_{11}, etc. are the direction cosines of the principal axes of the particles with respect to OX, OY and OZ. Since the external field is assumed to be applied along OY and propagation direction is along OZ

$$C_{21} = \cos\theta, C_{11} = \sin\theta\cos\varphi$$

'θ' Denotes the polar angle made by the axis of the nanomagnetic particle with respect to the direction of the magnetic field and 'ϕ' is the azimuthal angle made by the axis with respect to OX. The elements of the resultant scattering matrix then obtained by weighing the elements P_1, etc. with the orientation distribution function $f(\theta, \phi)$ and integrating over the angle θ and ϕ

$$\int\int P_1 f(\theta,\phi) d\theta d\phi = \int_0^{2\pi}\int_0^{2\pi} \alpha_0(\sin^2\theta\cos^2\phi) f(\theta,\phi) d\theta d\phi \quad (11)$$

A similar equation holds for P_2. The orientation distribution function $f(\theta,\phi)$ is given by

$$f(\theta,\phi) = \frac{N\exp\left(\frac{-U}{k_B T}\right)}{\int_0^{2\pi}\int_0^{2\pi}\exp\left(\frac{-U}{k_B T}\right)\sin\theta d\theta d\phi} \quad (12)$$

U is the potential energy of the particles, k_B is the Boltzmann constant and T is absolute temperature. If the dipolar magnetic moment of the particles along their symmetry axis is m then $U = -\mathrm{mH}\cos\theta$. Since the torque due to anisometry (shape anisotropy) of the particles is very weak its contribution may be neglected. Integration of Eq. (11) yields

$$\int\int P_1 f(\theta,\phi) d\theta d\phi = N\left[\frac{\alpha_o + \alpha_e}{2} - \frac{\alpha_o - \alpha_e}{2} L(h)\right] = P(h) \quad (13)$$

$$h = mH/k_B T \text{ and } L(h) = 1 - \frac{2}{h}\left(\coth\ h - \frac{1}{h}\right) \quad (14)$$

The dipolar moment $m = m_s V$. Eqs. (4)–(7) with the above equations will be transformed to

$$(C_{ext})_l = -4\pi kN\ \mathrm{Im}[\alpha_e + (\alpha_e - \alpha_o)L(h)] \quad (15)$$

$$(C_{ext})_r = -4\pi kN\ \mathrm{Im}\left[\frac{(\alpha_e) + (\alpha_o)}{2} - \left(\frac{(\alpha_e) - (\alpha_o)}{2}\right)L(h)\right] \quad (16)$$

$$n_l - n_r = 2\pi kN\ \mathrm{Re}\left(\frac{(\alpha_e) - (\alpha_o)}{2}\right)[3L(h) - 1] \quad (17)$$

$$(C_{\mathrm{ext}})_K = (C_{\mathrm{ext}})_r \quad (18)$$

In absence of the magnetic field, all the extinction cross sections reduces to

$$(C_{\mathrm{ext}})_0 = -4\pi kN\ \mathrm{Im}(2\alpha_e + \alpha_o)/3 \quad (19)$$

For experimental analysis it is convenient to determine parameters which are independent of concentration of the fluid. We define

$$Q_L = \frac{(C_{ext})_l}{(C_{ext})_0} = (Q_L)_\infty + \frac{3\mathrm{Im}(\alpha_e - \alpha_o)}{(\alpha_e + 2\alpha_o)}L(h) \quad (20)$$

$$Q_R = (Q_R)_\infty - \frac{1}{2}[Q_L - (Q_L)_\infty] \quad (21)$$

$$(Q_L)_\infty = 2(Q_R)_\infty = \frac{3\mathrm{Im}(\alpha_e - \alpha_o)}{(\alpha_e + 2\alpha_o)} \quad (22)$$

Q_τ (τ may be L or R) are determined from measurements of transmitted intensity in zero as well as applied field. If (ΔI_H) is the magnetically induced change in intensity and I_0 is the intensity in zero field and I_i is the intensity of incident light then

$$Q_\tau = 1 - \frac{\ln\left\{1 + \frac{\Delta I_H}{I_0}\right\}}{\ln\left(\frac{I_0}{I_i}\right)} \quad (23)$$

The extinction coefficients of a ferrofluid under a magnetic field satisfies the following relations [38]

$$\begin{aligned} &(i) \quad Q_K = Q_R \\ &(ii) \quad Q_L - 1 = 2(1 - Q_R) \end{aligned} \quad (24)$$

In the above relations Q_K is the extinction coefficient in the longitudinal configuration. The range of validity of the above relations may be extended to Rayleigh-Gans region when particles are absorbing [42,43]. In summary, dipolar scattering theory can explain observed magneto-birefringence as well as magneto-dichroism in a ferrofluid. In earlier papers we have applied the above criteria to confirm that each individual nanomagnet contribute to the observed effects and experimentally determined size and saturation magnetization agree well with that determined by other methods [19,32,35]. Analysis of observed effects may become complicated and confusing when investigations are carried out only in longitudinal configuration i.e. when direction of propagation of light and the direction of applied field are parallel (measurement of Q_K only) [34]. These authors have reported variation of transmitted intensity of light from a ferrofluid as a function of field and observed a minimum in it. Further they have found that effect is independent of the state of polarization. They have analyzed the data on basis of chain formation and using Mie theory. Chains have cylindrical symmetry and Mie theory is for spherical particle. Reported observation of Faraday rotation is also intriguing. Rotation of plane of polarization of light by a system in longitudinal field configuration is known as Faraday rotation [44–46]. As per the dipole scattering theory the propagation in this configuration should be scalar and Faraday rotation should not be observed. This conclusion was verified in our experimental work on well stabilized magnetic fluids [19,43]. Hence, to arrive at non-ambiguous inference it is necessary to study effect in transverse configuration. If a long chain is formed then, in transverse configuration a distinct diffraction line will be observed in direction normal to both the direction of incident light and the direction of applied magnetic field [33,47]. In earlier papers we have shown that if dispersion contained a mixture of nanomagnetic as well as micron sized magnetite particles then it will exhibit minimum in transmission as well as forward scattering pattern [48,49]. If the ferrofluids used in Ref. [34] contained both nanomagnetic particles as well as a small number of field induced large aggregates then also observed effects (scattering in forward direction and a minimum) can be accounted on the line of above theory .In short measurements in transverse configuration are helpful to ascertain formation of chains in a ferrofluid.

3.3. Mixture of ferrofluid and suspension of micron sized diamagnetic particles

We consider a suspension of ferrofluid and suspension of micron sized diamagnetic particles having a large anisotropic diamagnetic susceptibility e.g. disc shaped graphite particles. It is known that a non magnetic inclusion in otherwise uniform magnetized medium mimics a magnetic *hole* [50,51]. Optical extinction due to such holes in ferrofluids subjected a static uniform magnetic field is not studied. Theoretical expressions for normalized extinction parameters for such a system are derived and a simple experimental technique to separate out the effects due to such holes is described.

It is shown that a ferrofluids under the influence of static uniform magnetic field behaves like a quasi-homogeneous continuum [1]. When nonmagnetic particles are introduced in the fluid, the particles mimics magnetic holes in otherwise uniformly magnetized medium. These holes acquire induced diamagnetic moment equal to the magnetic moment of the ferrofluid they displaces. Skjeltorp [50] has shown that several condensed matter phenomena like melting; crystallization, phase transition, etc. can be simulated in such a system. The dynamics of such holes are also found to be interesting [52]. It has been shown that in rotating magnetic field holes rotate in opposite direction to that of the field. Usually spherical particles are used for such study. Earlier we have studied magnetically induced optical effects in such systems [32]. If instead of spheres ellipsoidal particles are used then over and above magnetically induced effective diamagnetic susceptibility of the holes, particles own anisotropic crystalline susceptibility will also contribute to the net effect. Consequently magneto optical effects in such a suspension will be governed by three types of torques (i) the torque due to permanent dipolar magnetic moment of nanomagnetic particles of the ferrofluids, (ii) the torque due to magneto crystalline anisotropy of the diamagnetic particles and (iii) the torque due to the effective diamagnetic anisotropy of the holes. Again the torque due to shape anisotropy being small may be neglected. In what follows we shall derive expressions of normalized magneto-optical extinction coefficients (Q_H). As in previous case we shall assume that single and independent scattering theory is applicable in the case. The nonmagnetic ellipsoidal particles are assumed to be very large compared to the wavelength of the incident light. This leads to extinction coefficient equal to twice the geometrical area $\hat{A}$ of shadow the particles [39]. When N′ particles are oriented under a magnetic field the total projected area of N′ particles per unit volume will be different in different field directions. Hence, $C^T = 2\hat{A}N'(\phi'_h)^T$, $C^L = 2\hat{A}N'(\phi'_h)^L$, T and L respectively represent transverse and longitudinal configuration of the field direction with respect to the direction of propagation. The functions $(\phi'_h)^{T,L}$ were derived earlier [39] for diamagnetic particles and are given by

$$(\phi'_h)^T = \frac{4h'}{\pi E_h}\int_0^{\pi} \exp(-h'^2\cos^2\theta)\sin^2\theta d\theta \tag{25}$$

$$(\phi'_h)^L = \frac{1}{h'E_h}[1-\exp(-h'^2)] \tag{26}$$

$$h' = \sqrt{\frac{V(\chi_2-\chi_1)H^2}{k_B T}} \tag{27}$$

In addition to the above we also have now effective magnetic susceptibility due holes whose magnetic moment will be given by $M_V = -MV$, where V is the volume of the displace fluid. The effective potential energy due to holes is given by [51]

$$U_{holes} = -\frac{(\chi_{holes}VH^2)}{a^3k_BT}(1-3\cos^2\theta) \tag{28}$$

The net potential energy will be

$$(U) = U_{nanomagnetic} + U_{diamagnetic} + U_{holes} \tag{29}$$

Following the procedure given in case of the derivation of extinction parameters for a ferrofluid, expressions for Q_F are obtained [5]. The governing function now assumes the form

$$(\varepsilon_h)_\tau = \frac{3}{1+R}[\phi(h')_d + R\{\phi(h)_n + \phi(h'')_{holes}\}] \tag{30}$$

$$\phi(h'')_{holes} = \frac{1}{2\pi}\exp[-3h''\cos^2\theta]\left[1-h''+\frac{9}{10}(h'')^2\right] \tag{31}$$

$$h'' = \sqrt{\frac{\chi_{holes}VH^2}{a^3k_BT}} \tag{32}$$

$\phi(h)_n = L(h)$ is the Langevin function defined earlier. The factor R depends on the ratio of number of the nanomagnetic and diamagnetic particles as well as on ratio of optical polarizability of nano and diamagnetic particles respectively.

$$R = \frac{N_{nano}}{N_{diam}}\mathrm{Im}\left(\frac{\alpha_e-\alpha_o}{\alpha'_e-\alpha'_o}\right) \tag{33}$$

Plots of $\varepsilon(h)_\tau$ *versus* the composite field parameter h for different values of R are shown in Fig. 1. It is observed that when R is between 1.6 and 0.8 a minimum in the function appears and it shifts to higher value of $h(H)$ when R decreases from 1.6 to 0.8. Graphite particles possess a large diamagnetic anisotropy as well are disc shaped. When a magnetic field is applied to a dispersion of micron sized graphite particles, field and polarization dependent extinction of light is observed [53,54]. In an earlier paper magneto optical extinction was also studied for a mixture of ferrofluids and graphite dispersion [32]. But effect of holes on observed parameters was neglected. An attempt was made by Trivedi to study contribution of holes in such a mixture [55]. A commercial hydrocarbon based ferrofluid FN40 and oleic acid coated graphite dispersion having average size of 3 µm was selected for the purpose. Two suspensions having weight concentration 0.57 mg/ml, 1.47 mg/ml of graphite particles were mixed with a diluted sample of the ferrofluid. Experimental set up was similar to that described elsewhere [39]. Variations of Q_τ with the applied field H, respectively for the diluted ferrofluid and the graphite suspension (0.57 mg/ml) are shown in Figs. 2 and 3. A ferrofluid with nanomagnetic particles having same size of the particles (a monodispersed system) does not show any minima or maxima in variation of Q_τ with magnetic field [19]. If, a fluid contains a small number of large sized magnetic particles (aggregates) then a minimum in Q_L is observed [39,48]. Hence, observed Minimum in Q_R in Fig. 2 is attributed to the presence of small number of aggregates in the ferrofluid. Continuous lines in Fig. 2 is generated theoretically using observed values of the filed corresponding to H_{min} = 0.009T and H_{max} = 0.0225T and R = 0.07. It is observed that extremum is observed only in case of variation of Q_L with field. Similarly, in Fig. 3 lines are drawn by inserting the values of $(\chi_e - \chi_o) = -4.0 \times 10$–10 emu/kg and particle size = 0.8 µm. Figs. 4a and b show variation of Q_τ with H for mixtures of the ferrofluid and suspensions of 0.57 mg/ml and

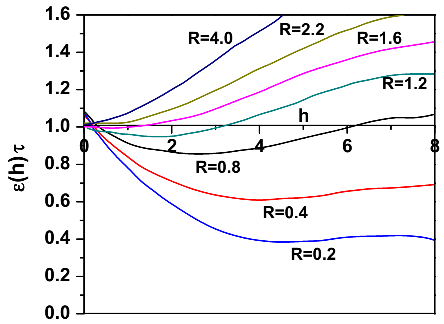

Fig. 1. (Mehta): variation of the orientation distribution function $\varepsilon(h)_\tau$ with 'h' for different values of R.

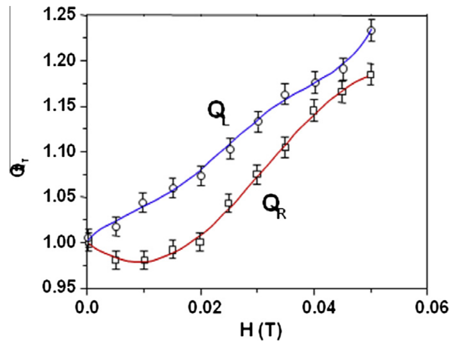

Fig. 2. (Mehta): variation of extinction coefficients Q_L and Q_R of the ferrofluid with applied field.

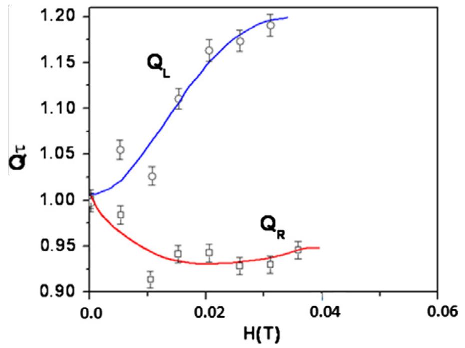

Fig. 3. (Mehta): variation of $Q_\tau(\tau = L, R)$ with field for graphite dispersion.

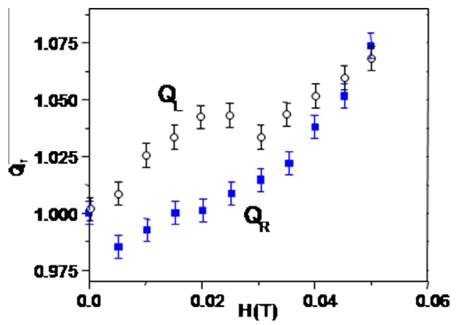

Fig. 4a. (Mehta): variation of $Q_\tau(\tau = L, R)$ with the field H for mixture of ferrofluid and graphite suspension having 0.57 mg/ml of graphite particles.

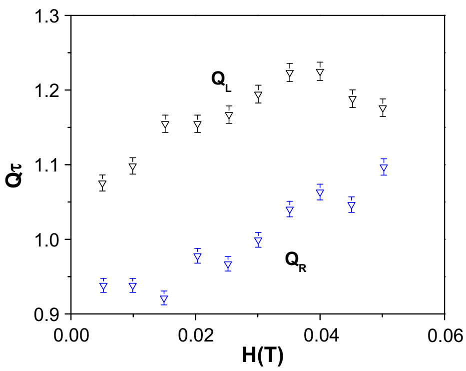

Fig. 4b. (Mehta): variation of $Q_\tau(\tau = L, R)$ with the field H (Tesla) for a mixture of ferrofluid and 1.47 mg/ml of graphite dispersion.

1.47 mg/ml graphite particles respectively. A distinct minimum is observed in Q_R while Q_L exhibits either a maximum or minimum. Similar results were obtained for a third sample (not shown). Results show that the field corresponding to Q_R minimum increases as number concentration of graphite particles increases. The later is proportional to R. This behaviour is similar to that observed in the theoretical plots (Fig. 1). Curve fittings and interpretation of variation in Q_L could not be carried out since all the suspensions were polydispersed. More over thickness of the graphite particles were not negligible and to account for these, the above theory needs modifications.

4. Summary

We have shown that in order to arrive at an unambiguous conclusion regarding field induced optical extinction of a ferrofluids it is necessary to measure the polarization dependence of extinction of light in transverse configuration of applied magnetic field. If it depend on the direction of electric vector of the incident linearly polarized light, then one should also measure optical extinction in the longitudinal configuration. If the normalized extinction coefficients obey the relations (20) then it can be concluded that no aggregation (chain formation) is formed. If chain formation occurs then in transverse configuration a distinct diffraction line in direction perpendicular to the propagation direction of light will be observed. In case of presence of aggregates a minimum in Q_R as well as Q_K will be observed.

A mixture of ferrofluids and dispersion of micron sized anisotropic diamagnetic particles may exhibit a minimum in Q_R depending on ratio of number of nanomagnetic particles in the ferrofluids and number of the micron sized particles. Contribution of 'magnetic holes' can be inferred from the measured variation of $Q_\tau(\tau = L, R)$ if the scattering sample is fairly monodispersed and thickness of the large particles is negligible. To account for polydispersed system and finite thickness of larger particles the theory developed in this paper is required to be modified.

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