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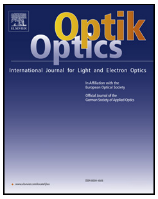

Original research article

Extinction of light by a Ferrocell and ferrofluid layers: A comparison

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ABSTRACT

Recently a magneto-optical device called Ferrocell has become commercially available. As per manufacturer data, it mainly consists of a ferrofluid layer sandwiched between two glass plates, somewhat similar to a Hele-Shaw cell. Optical transmission changes with applied magnetic fields are not studied in detail for this device. We investigated transmission changes for one such commercial device and compared it with ferrofluid filled in a thin glass cuvette cell. Results show that the ferrofluid contained in the Ferrocell consists of a mixture of small and large particles.

1. Introduction

Ferrofluid is a versatile material. It exhibits several novel and interesting phenomena in the presence of externally applied magnetic fields, like defying gravity, levitation and spike formation. Its physical properties are also modified by magnetic fields and a number of applications in many branches of science and technology are made possible by the characteristics of this fluid. Ferrohydrodynamics- the fluid mechanics of ferrofluids was developed to explain the salient features of the fluid. Research areas and applications of ferrofluids seem to be an ever-growing topic. In the recent past, their applications in optical and photonic devices have drawn considerable interest [1–6]. Even before the availability of ferrofluids, several papers were published on magneto-optics of magnetic colloids [7–11]. Commercially available ferrofluids and synthesis of the fluids with different materials and the different medium has made it possible to explore novel applications like polarization modulator, optical switches, filters, photonic band-gap materials, ferrolens, etc. [12–16]. In earlier works, a stable and water-based dilute dispersion of monodomain magnetite particles was mainly used and stabilization was achieved by charge balancing on each particle, while in ferrofluids, a well stabilized dispersion of magnetic nanoparticles in any liquid medium is used. Stabilization is mostly achieved either by steric and Coulomb repulsion. The saturation magnetization of ferrofluids is much higher than those used in earlier investigations. Such a fluid under normal condition is opaque to visible radiation, but when a thin film of this fluid is placed in a very short optical path it will be semi transparent. Optical properties under such a transparent system can be investigated when subjected to a variable magnetic field. Thin films of ferrofluid are now available commercially under the name of Ferrocell. A ferrocell contains a non-aqueous ferrofluid sandwiched between two glass plates and sealed from all the sides. The fluid layer is < 15 microns thick. When placed under a strong magnetic field of a rare earth magnet it can function like a lens, grating or polarization modulator [17–22]. These devices are based on magnetically induced optical anisotropy in the fluid. The difference between a usual ferrofluid filled Hele-Show cell and the commercially available Ferrocell is that in the later ferrofluid has no free surface, hence its movement as a whole is restricted and the former may be a spectrometer cuvette with very short path length. It will be interesting to compare induced optical anisotropy in both cells.

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Uses of suitable liquids in optical devices like a camera lens and a microscope objective are well known. Both reflective and refractive devices are possible with such liquids. Reflective devices are mainly variable mirrors and are used in telescopes. Similarly, refractive devices are based on tuning the refractive index of the liquid by varying its temperature. Advantages are no moveable parts, a very large range of variable focus, smaller size, and flexibility. The surface shape can usually be changed by applying an electric field. In mobile phone cameras, endoscopes, etc., this technique is used [23–29]. Here, the focus of the lens is controlled by changing the shape of the surface of the liquid. Water is the most convenient choice for this purpose as it forms bubble shape when adhered to glass or plastics. Another liquid like oil is mixed with this water and the mixture is sandwiched between two transparent glass (plastics) flat plates. The focal length of such a lens is decided by the surface profiles of the liquids. The electric field is used to vary the profiles [29].

Ferrocell also is known as Ferrolens, a fluid, in which a magnetic field is used to vary the refractive index of the fluid. The fluid is composed of magnetic nanoparticles stably suspended in a host liquid. Such a lens has several possible applications [17,21]. Ferrofluid sealed in such a lens is usually commercial ferrofluids. Till now comparison of magnetically induced effects of magnetic fluid sealed in a ferrolens and those placed in an optical cell having at least one free surface are not reported. We found that there are certain significant differences between the two. The present paper aims to describe the comparison of the two magneto-optical effects and their implications.

2. Experimental

2.1. Sample preparation

Ferrofluid was synthesized using a well-known co-precipitation technique [30]. Stoichiometric solutions of GPR grade $FeCl_3$ $6H_20$ and $FeSO_4$ $7H_20$ were dissolved in deionized water and were mixed together. This will result in free Fe^{3+} and Fe^{2+} ions in an aqueous solution. Ammonia solution (8 M NH_4OH) was then added to the above mixture while thoroughly stirring. This redox reaction was carried at room temperature under constant stirring. Nearly 2 mL of Oleic acid was then added to the solution and heated at 95 °C for 5 min and cooled to room temperature. After decantation, the product was washed a number of times with hot distilled water and finally washed with acetone. This acetone wet slurry was dispersed in kerosene and stirred. The fluid was heated to 60 °C, so as to remove acetone and the final product was centrifuged at 12,000 rpm for 10 min and impurities and aggregates were removed by decanting the supernatant. Domain magnetization, saturation magnetization, and average particle sizes were measured by usual techniques and were found to be $M_d = 320$ emu/cc, $M_s = 66$ G and $d = 10$ nm (Fig. 1(b)), respectively. The original fluid having density $\rho = 0.92$ gm/cc was diluted to avoid multiple scattering. Four different concentrations 5%, 10 %, 15 % and 20 % of the above fluid were used in the present study.

Ferrocell was gifted by Timm A. Vanderelli, of Ferrocell USA for our investigations. As per the manufacturer's data, the fluid used in the Ferrocells was EFH 1 which was obtained from Ferrotec Corp. USA [20]. It is a stable dispersion of nanomagnetic particles in mineral oil. As per the technical data of the ferrotech corporation, it has a saturation magnetization of 44 m T and 6 cP viscosity. The ferrofluid layer of less than 15 microns was sandwiched between two glass plates. The layer was surrounded by immiscible oil and sealed on all sides (Fig. 1(a)).

2.2. Experimental setup

The experimental set up is depicted in Fig. 1(c). The optical setup consists of a linearly polarized He-Ne laser with a wavelength of 633 nm and 10 mW output power (Optochem International). A polarizer was used to orient the direction of polarization with respect to the direction of the field. The magnetic field was either generated by electromagnets with constant current variable power supply or permanent rare earth magnets (in case of the Ferrocell). The Sample was placed between the poles of the electromagnets. The ferrofluid sample was filled in a 1 mm path length of glass cuvette. The direction of propagation of incident light was kept transverse to the direction of the magnetic field. Output intensity was detected by a photodetector and recorded on a nanoameter. Measurements were carried out in two orthogonal states of polarization with respect to the direction of the applied field. In one case the E-vector of the incident light was oriented parallel to the direction of the field and in the other was oriented to the perpendicular the field.

3. Magneto-optical transmission

The transmission changes of light passing through a colloidal solution under an influence of the magnetic field depend on field-induced anisotropy of the system. The latter depends on the size shape, optical and magnetic properties of the dispersed particles. The induced optical anisotropies observed in systems with small and large particles are markedly different. If the particles are sufficiently small compared to the wavelength of the incident light, dipole scattering theory can be used to explain the transmission changes, if the particles are large compared to the wavelength of the incident light, geometrical optics can be used to explain such changes. When the system contains particles of intermediate sizes or when it contains small as well as large particles complications arise. [31–33]. It was shown that in the case of a dipole scattering by anisotropic particles, the extinction coefficient under the influence of a magnetic field depends on the polarization vector of the incident plane-polarized light [31]. For larger particles, the extinction depends on the geometrical shadow of the particles and for the mixture of both the types of particle, extinction depends on the ratio of large to small particles [33]. Comparing the experimental data with the theoretical one it is convenient to express the transmission changes as the ratio of the field-dependent extinction coefficient (C_F) and the extinction coefficient in zero fields (C_0) as follows

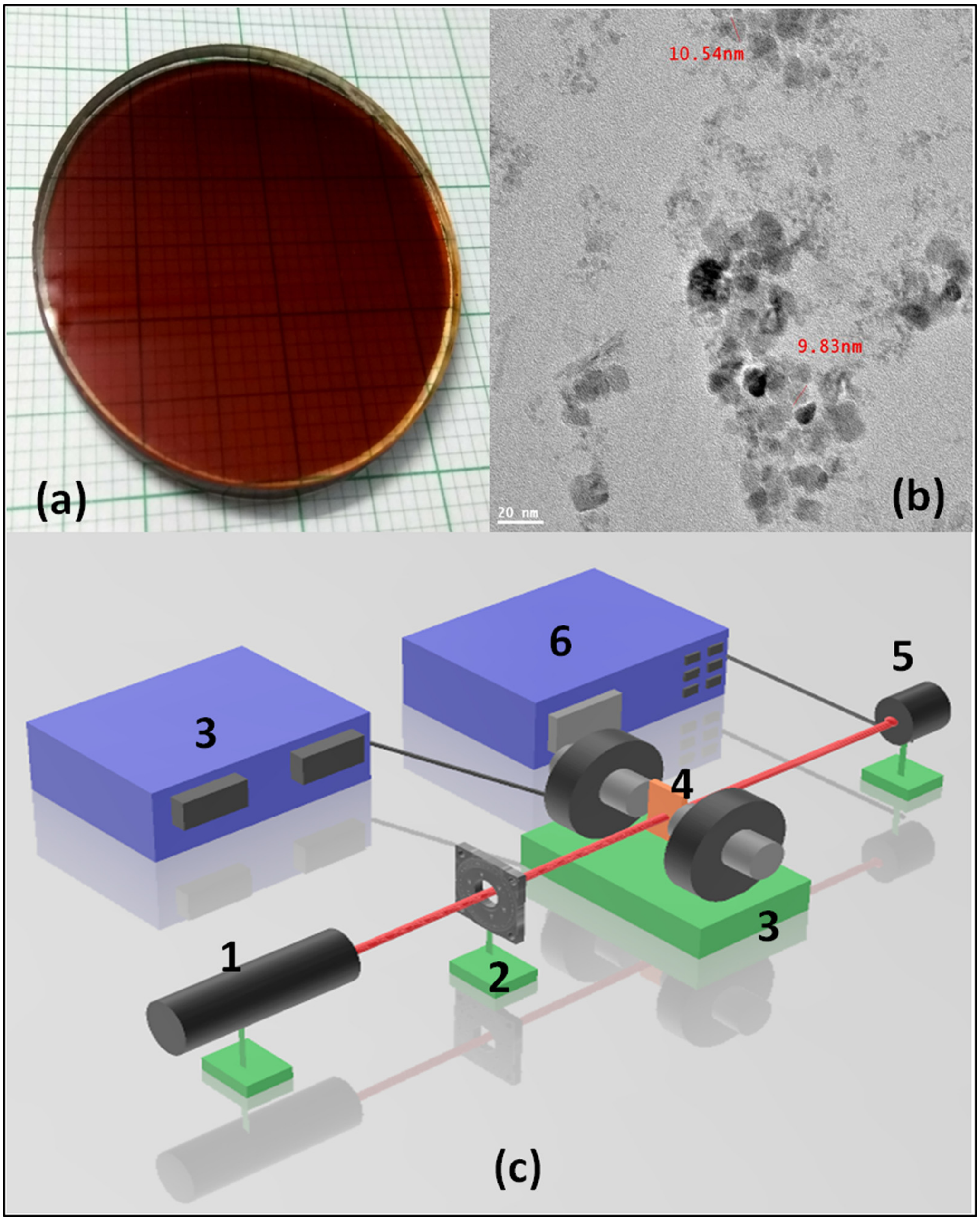

Fig. 1. (a) Image of ferrocell (b) TEM image of synthesized Fe_3O_4 particles (c) Experimental setup 1) plane-polarized He-Ne Laser, (2) polarizer, (3) electromagnet & power supply, (4) sample, (5) detector, (6) Nanoameter.

$$Q_F = \frac{C_F}{C_0} = 1 - \frac{\ln\left(1 + \frac{\Delta I}{I_0}\right)}{C_0} \tag{C.1}$$

Q_F, C_F, and C_0 are respectively ratios of extinction parameters in a magnetic field, extinction coefficients in the field and zero fields. ΔI and I_0 are changes in intensity in the field and intensity in the zero-field, respectively, 'ln' indicates natural logarithm [34]. Further, for dipole scattering following relations are also almost holds.

$$Q_{\parallel} - 1 = 2(1 - Q_{\perp}) \tag{C.2}$$

$$Q_{\perp} = -Q_k \tag{C.3}$$

$Q_{\parallel}$, $Q_{\perp}$, and Q_k are respectively extinction parameters for a plane-polarized light transmitted with its E-vector parallel and perpendicular to the direction of the magnetic field when the incident light is propagating in the direction perpendicular to and Q_k for light propagating along the direction of the field. In the latter case, extinction is independent of the state of polarization of light [32]. Further, it was also shown that when the fluid consists of a large number of such particles the extinction coefficients should be weighted by an orientation distribution function as well as a size distribution function. Without going into detailed derivation described in Ref [33]. we summarized the results for the particles with a permanent magnetic moment that is in our case magnetite particles. It is assumed that the conditions of single and independent scattering are followed by the fluid.

$$C_{\parallel} = -4\pi kN\,[\,\mathrm{Im}\,\alpha_2 + \mathrm{Im}\,(\alpha_1 - \alpha_2)\phi(h)] \tag{C.4}$$

$$C_{\perp} = -4\pi kN\left[\mathrm{Im}\left(\frac{\alpha_1 + \alpha_2}{2}\right) - \mathrm{Im}\left(\frac{\alpha_1 - \alpha_2}{2}\right)\phi(h)\right] \tag{C.5}$$

$$C_0 = -4\pi kN\left[\frac{2\alpha_2 + \alpha_1}{3}\right] \tag{C.6}$$

Where, $\phi(h) = 1 - \frac{2}{h}\left(\coth - \frac{1}{h}\right)$; $h = \frac{mH}{k_B T}$ and $m = I_s V$

In the above expressions α_j (j = 1,2,3) are the components of polarizability tensor, k is propagation vector, V is the volume of the particles, I_s is the saturation magnetization, $\mathrm{k_B}$ is Boltzmann constant, and T is absolute temperature. Assumed to be axially symmetric hence $\alpha_2 = \alpha_3$; N is the number of particles in a unit.

In the case of very large particles as compared to the wavelength of light in the medium, the extinction coefficient has been shown to be twice the geometrical area of the shadow of the particles [[35], p. 107]. When a large number of such particles, each having area 'a' are oriented under a magnetic field, the total projected area of N' particles per milliliter will be different in different field directions. This leads to

$$C_T = 2(G_H)T \tag{C.7}$$

Where subscript T refers to the transverse configuration, $\mathrm{G_H}$ depends upon the magnetic properties of the particles and their shape. If 'a' is the area of each particle

$$C_T = aN'\phi'(h)_T \tag{C.8}$$

For polar particles the function $\phi'(h)_T$ is given by the following expressions:

$$\phi'(h)_T = \frac{4h}{\pi(e^h - 1)}\int_0^{\pi/2}\{\exp(h\cos\gamma)\}\sin^2\gamma d\gamma \tag{C.9}$$

γ is the angle made by the particle w.r.t. the field direction.

In the case of a ferrofluid containing a mixture of very fine and very large particles and assuming the conditions as given in the reference [33], we obtain

$$(C_{Mixture})_T = (C_{Fine})_T + (C_{L\arg e})_T \tag{C.10}$$

In the transverse configuration $\mathrm{C_{Fine}}$ depends on the orientation of the electric vector of the incident linearly polarized light with respect to the direction of the field. Hence, the extinction coefficients of the mixture can be expressed as

$$(C_M)_T = C_{\parallel,\perp} + C_T \tag{C.11}$$

With the above expressions of C_T and $C_{\parallel,\perp}$ we obtain

$$(C_M)_T = [A + B\phi(h)]N + aN'\phi'(h)_T \tag{C.12}$$

Here, $A = -4\pi kN\,\mathrm{Im}\left(\frac{\alpha_1+\alpha_2}{2}\right)$ and $B = 4\pi k\,\mathrm{Im}\left(\frac{\alpha_1-\alpha_2}{2}\right)$

From the above expression, it can be inferred that the extinction coefficient, in this case also, depends on the ratio of fine to very large particles (R = aN′/BN). In the absence of the applied field, we get

$$\frac{C_0}{NB} = \frac{A}{B} + \frac{1}{3} + R \tag{C.13}$$

Combination of the last two-equation yields

$$\frac{(C_M)_T - AN}{C_0 - AN} - \frac{\phi(h) + R\phi'(h)_T}{\frac{1}{3} + R} = M(h)_T \tag{C.14}$$

This function $M(h)_T$ for the mixed system describes the variation of extinction with respect to the field strength when the light beam propagates transversely to the magnetic field with its electric vector perpendicular to the field. Similarly, for transverse propagation with the electric vector parallel to the magnetic field

$$M(h)_{\parallel} = \frac{2\phi(h) - R\phi'(h)_T}{\frac{2}{3} - R} = \frac{DN - 2(C_M)_{\parallel}}{DN - C_0} \tag{C.15}$$

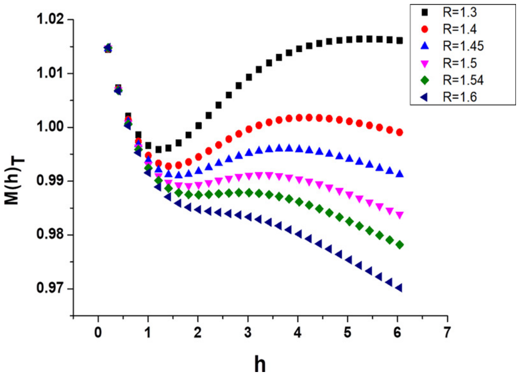

Fig. 2. Characteristic function $M(h)_T$ for different values of R.

Plots of these functions are discussed in more detail in ref [33,36]. It was deducted that when R>1 then the extinction first decreases with an increase in field, reaches a minimum, and then increases to above its zero-field value. Fig. 2 shows the behavior of function $M(h)_T$ with a change in R.

4. Results and Discussion

4.1. Ferrocell

Fig. 3 shows the extinction parameters for the Ferrocell. It is observed that extinction parameters are not significantly different for the two states of polarization vectors and also almost independent of the field. It seems that only with a small increase in applied field, stable aggregates are formed and there is little change in chain length of the aggregates with an increase in the field.

Fig. 4 shows the matching of experimental values of Q_F for different field H with the theoretical curve of $M(h)_T$ for R = 1.7 and for a diameter of single-particle 14 nm. These findings suggest the average size of the particles in the Ferrocell is large and there may be a very small number of the fine single particles. It may be remarked here that original ferrofluid in the ferrocell may have fine single particles but after exposure of high field large irreversible chains could have been formed.

4.2. Ferrofluid

Fig. 5 shows variations of field-induced scattering parameters with applied static magnetic fields in the case of the ferrofluid samples. The following conclusions are drawn from these variations.

(1) The dipole scattering criterion (Eq. (C.2)) is not obeyed for all the dilutions. This is surprising because in the original fluid average size of the particles was found to be 10 nm. (Fig. 1(b))
(2) Changes in the variations are observed to be larger for lower dilutions.
(3) The magnetic field induced extinction of light for both the state of polarization decreses with increasing field.
(4) $Q_{\parallel} > Q_{\perp}$. Hence, the samples do not contain only large particles

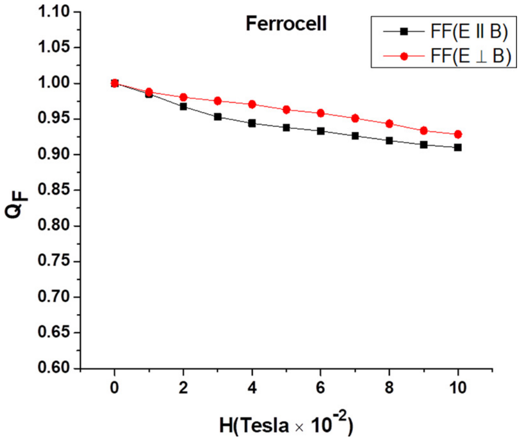

Fig. 3. Magnetically induced extinction of light in ferrocell, the symbol ● represents data for E⊥H and symbol ■ represents data for E||H.

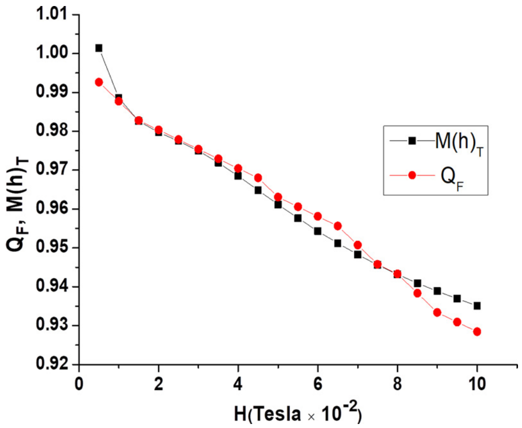

Fig. 4. Fitting of Experimental Q_F with theoretical $M(h)_T$ (R = 1.7).

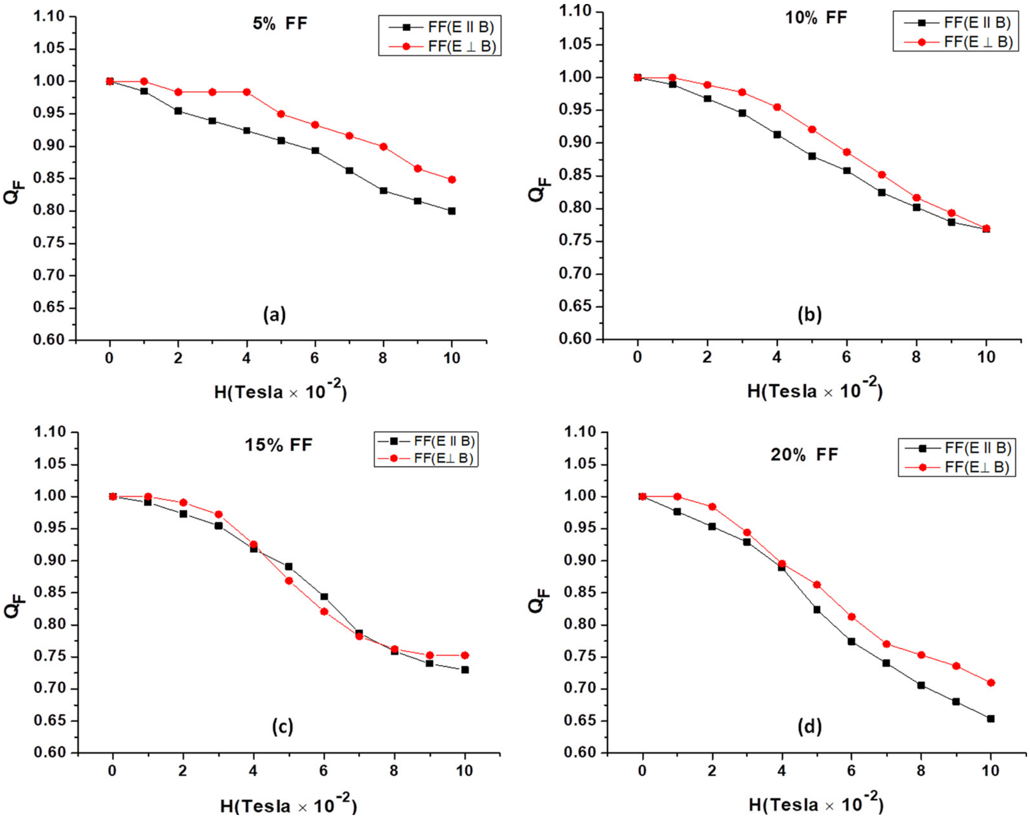

Fig. 5. The magnetically induced extinction of light in different concentration of ferrofluid, (a) 5% FF, (b) 10 % FF, (c) 15 % FF, (d) 20 % FF. A symbol ■ represents data for E||H and the symbol ● represents data for E⊥H.

(5) Fluctuations in variations of extinction also related with dilution.

From the above observations and the theoretical considerations described earlier we can infer the following:

All the samples contain small as well as large particles. The proportions of these changes with dilutions. In other words, the fluid is dilution sensitive. Such aggregates in the water-based fluid were also earlier observed [11,36]. In such a dynamic system, the size of some particles may be in the intermediate range. For such particles, theoretical expressions described earlier are not valid [31,32]. Further, the ratio of large to fine particles also changes with time and field. Hence, no theoretical comparison is carried out.

5. Conclusion

Extinction parameters for a commercially available Ferrocell and that of diluted samples of synthesized ferrofluid were determined. From the theoretical considerations, it is deducted that all the ferrofluid samples, as well as the Ferrocell, contains a mixture of small and large particles. These aggregates are field-induced and are in the shape of liner chains. The factor R which depends on the ratio of a number of large and fine particles for the Ferrocell is 1.7. For the diluted samples, it was inferred that the fluid is dilution sensitive and the size of aggregates changes with the field. In the case of the Ferrocell, extinction is almost independent of the state of polarization of the incident light and remains almost independent of the applied field.

Declaration of Competing Interest

The authors declare no conflicts of interest.

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References

[1] D. Jamon, et al., Ferrofluids, sol-gel method and materials for optical applications, Indian J. of engineering & materials sciences 11 (2004) 237–240.
[2] Qiuling Chen, et al., Plasmon enhanced faraday rotation in Fe 3 O 4/Ag ferrofluids for magneto-optical sensing applications, Plasmonics 13 (2018) 353–363, https://doi.org/10.1007/s11468-017-0606-1.
[3] John Philip, Junaid M. Laskar, Optical properties and applications of ferrofluids—a review, J. Nanofluids 1 (2012) 3–20, https://doi.org/10.1166/jon.2012.1002.
[4] Chintamani Pai, M. Shalini, S. Radha, Transient optical phenomenon in ferrofluids, Procedia Eng. 76 (2014) 74–79, https://doi.org/10.1016/j.proeng.2013.09.250.
[5] Daniel Homa, Gary Pickrell, Magnetic sensing with ferrofluid and fiber optic connectors, Sensors 14 (2014) 3891–3896, https://doi.org/10.3390/s140303891.
[6] Rajesh Patel, Rasbindu V. Mehta, Ferrodispersion: a promising candidate for an optical capacitor, Appl. Opt. 50 (2011) G17–G22, https://doi.org/10.1364/AO.50.000G17.
[7] Q. Majorana, Sulla Birifrangenza Magnetica e su altri Fenomeni che l'accompagnano, Rend. Accad. Lincei 11 (1902) 531.
[8] C.W. Heaps, Optical and magnetic properties of a magnetite suspension, Phys. Rev. 57.6 (1940) 528, https://doi.org/10.1103/PhysRev.57.528.
[9] W.C. Elmore, Theory of the optical and magnetic properties of ferromagnetic suspensions, Phys. Rev. 60 (1941) 593, https://doi.org/10.1103/PhysRev.60.593.
[10] Hans Mueller, Morris H. Shamos, Magneto-optical properties of ferromagnetic suspensions, Phys. Rev. 61 (10) (1942) 631, https://doi.org/10.1103/PhysRev.61.631.
[11] M.J. Dave, et al., Optical transmission and birefringence of colloidal Fe3O4 in a magnetic field, Indian J. Pure Appl. Phys. 6.7 (1968) 364.
[12] Guojun Yu, et al., Tunable one-dimensional photonic crystals based on magnetic fluids, Opt. – Int. J. Light Electron. Opt. 124 (2013) 2713–2715, https://doi.org/10.1016/j.ijleo.2012.08.086.
[13] C.Z. Fan, G. Wang, J.P. Huang, Magnetocontrollable photonic crystals based on colloidal ferrofluids, J. Appl. Phys. 103 (2008) 094107.
[14] P. Agruzov, I. Pleshakov, E. Bibik, S. Stepanov, A. Shamray, Fast magneto-optical modulation in optofluidic devices based on ferrofluid- filled microstructured optical fibers, 2015 European Conference on Lasers and Electro-Optics - European Quantum Electronics Conference, Optical Society of America, 2015 paper CK_P_13.
[15] Yinping Miao, et al., Multidimensional microstructured photonic device based on all-solid waveguide array fiber and magnetic fluid, Nanophotonics 6 (2017) 357.
[16] Tianjun Yao, et al., Investigation of optical force on magnetic nanoparticles with magnetic-fluid-filled Fabry-Perot interferometer, Sci. Rep. 8 (2018) 12352.
[17] Alberto Tufaile, Timm A. Vanderelli, Adriana Pedrosa Biscaia Tufaile, Light polarization using ferrofluids and magnetic fields, Adv. Condens. Matter Phys. 2017 (2017).
[18] Alberto Tufaile, Timm A. Vanderelli, Adriana Pedrosa Biscaia Tufaile, Observing the jumping laser dogs, J. Appl. Math. Phys. 4 (2016) 1977–1988.
[19] A. Tufaile, T.A. Vanderelli, A.P.B. Tufaile, Light Polarization Using Ferrofluids and Magnetic Fields, Encontro de Física, Natal, Brazil, 2016 September.
[20] Timm Vanderelli of Ligonier, Pennsylvania. USPTO patent application #20070241745 Magnetic Flux Viewer. http://nanomagnetics.us.
[21] Emmanouil Markoulakis, Antonios Konstantaras, Emmanuel Antonidakis, The quantum field of a magnet shown by a nanomagnetic ferrolens, J. Magn. Magn. Mater. 466 (2018) 252–259.
[22] P. Domachuk, et al., Compact resonant integrated microfluidic refractometer, Appl. Phys. Lett. 88 (2006) 093513.
[23] Stefania Campopiano, et al., Microfluidic sensor based on integrated optical hollow waveguides, Opt. Lett. 29 (2004) 1894–1896.
[24] Pavel Polynkin, et al., Evanescent field-based optical fiber sensing device for measuring the refractive index of liquids in microfluidic channels, Opt. Lett. 30 (2005) 1273–1275.
[25] Fei Xu, Peter Horak, Gilberto Brambilla, Optical microfiber coil resonator refractometric sensor, Opt. Express 15 (2007) 7888–7893.
[26] Sergio Calixto, Martha Rosete-Aguilar, David Monzon-Hernandez, Vladimir P. Minkovich, Capillary refractometer integrated in a microfluidic configuration, Appl. Opt. 47 (2008) 843–848.
[27] Minghui Chen, et al., A miniaturized system for measurement of the refractive index of sub-microliter liquid, Chinese Opt. Lett. 17 (2019) 041201.
[28] Chao Liu, Di Wang, Qiong-Hua Wang, Variable aperture with graded attenuation combined with adjustable focal length lens, Opt. Express 27 (2019) 14075–14084.
[29] C.U. Murade, et al., Electrowetting driven optical switch and tunable aperture, Opt. Express 19 (2011) 15525–15531.
[30] Vishakha Dave, Rasbindu V. Mehta, Magnetooptical effect of immersion of nonmagnetic macroscopic objects in the ferrofluid, J. Nanofluids 5 (2016) 37–41.
[31] R.V. Mehta, H.S. Shah, M.J. Dave, Optical transmission through bentonite suspension in a magnetic field, J. Colloid Interface Sci. 35 (1971) 41–45.
[32] D.M. Bhagat, R.V. Mehta, H.S. Shah, Extinction of light by oriented large particles, J. Colloid Interface Sci. 81 (1981) 14–20.
[33] R. Mehta, et al., Magnetically induced extinction in a particulate system, IEEE Trans. Magn. 16 (1980) 1324–1331.
[34] R.V. Mehta, Optical properties of certain magnetic fluids, Thermomechanics of Magnetic Fluids (1977) 139–148.
[35] H.C. van de Hulst, Light Scattering by Small Particles, Wiley, New York, 1959.
[36] R.V. Mehta, et al., Magneto-optical effects in magnetic fluid containing large aggregates, J. Magn. Magn. Mater. 289 (2005) 36–38, https://doi.org/10.1016/j.jmmm.2004.11.011.