

Optical Properties of Fe_3O_4 Magnetic Fluid from Iron Sand

A Puspitaningrum¹, A Taufiq^{1,2}, A Hidayat^{1,2}, Sunaryono^{1,2}, N Hidayat^{1,2} and Samian³

¹Department of Physics, Faculty of Mathematics and Natural Sciences, Universitas Negeri Malang (State University of Malang), Jalan Semarang No 5, Malang 65145, Indonesia

²Center for Minerals and Advanced Materials, Faculty of Mathematics and Natural Sciences, Universitas Negeri Malang (State University of Malang), Jl. Semarang 5, Malang 65145, Indonesia

³Department of Physics, Faculty of Science and Technology, Universitas Airlangga, Jalan Sutorejo, Surabaya 60115, Indonesia

Email: arif.hidayat.fmipa@um.ac.id

Abstract: Nowadays, a high sensitive sensor for the magnetic field has become an essential tool that vastly desired in several fields, especially in biomedical application. Therefore, the development of preparing material for the magnetic sensor becomes crucial to be conducted. In this experiment, we propose the use of Fe_3O_4 magnetic fluid prepared from a local iron sand in Indonesia as a material for a magnetic sensor. In this work, optical activities of the Fe_3O_4 magnetic fluid as the effect of magneto-optics were performed under varying external magnetic field. The polarization direction change of the laser was detected as a function of the external magnetic field with the exponential function. Moreover, the intensity collected by a photodetector exhibited a linear correlation with the external magnetic field. These phenomena become strong evidence that the prepared Fe_3O_4 magnetic fluid opens potential to be applied further as sensors, especially as a high sensitive optics-based sensor for the magnetic field.

Keywords: Magnetic fluid, Fe_3O_4 , iron sand, polarization, magneto-optic.

1. Introduction

Magnetic fluid or often called as ferrofluid is a stable colloidal suspension consisting of magnetic nanoparticles with single-domain character and dispersing in a suitable liquid carrier [1]. The surfactant-based magnetic fluid has good stability broadening its applications in many fields. The applications cover such as for controlling lubricant migration [2], for antibacterial and anticancer [3], as a contrast agent for magnetic resonance imaging [4], for magnetic field sensor [1], and so forth. Furthermore, for application in a big amount, the development of synthesis method of local natural substance based magnetic fluid which is inexpensive and abundant is significant by using natural iron sand, for example.

Magnetic nanoparticle in the ferrofluid forms a colloid magnetic system caused by its permanent magnetic moment [5]. The distributed particles act as single-domain magnetic particles in the position



of random Brownian motion without the influence of external magnetic field. However, if it is exposed to an external magnetic field, the particles motions will be regular since they undergo a tensile strength along the field direction. It means that the dipolar interaction of the magnetic domains tends to increase. When the dipolar interaction becomes strong enough, the particles undergo the structure change [6-7]. First, the head-tail of the magnetic particles oriented randomly knits each other as long as the field direction by forming a number of chains. This chain-like structure is the result of the competition between the magnetic dipolar interaction and thermal interaction [8]. After that, these chains experience aggregation again through lateral smelting forming a bundle of the chain [9]. This phenomenon is known as zippering effect as well. Two effects happening simultaneously are interaction induced by thermal fluctuation and inhibitor of the local lateral field caused by the topology defect in the dipolar chain correlated to the type of this structure formation. As a consequence, there are many significant changes in the fluid characteristics observed. Regarding this case, specifically, the characteristics of magneto-optic and magneto-viscus are essential things and have attracted many experts' attention in the last years. Therefore, in this work, a study focused on the optical characteristic of Fe_3O_4 magnetic fluid detected by using magneto-optic effect is significant.

The magneto-optic effect is a symptom related to the light wave interaction with the external magnetic field applied in a material. The light polarization describes the vector orientation of electrical field from the light wave of a certain point in one period of vibration. In the light polarized circularly, the direction of propagation and the orientation of electrical field form either right and left round [10]. When the light wave propagates to penetrate the active optical material, the right and left polarization vector of the light wave moves with different velocity [11]. This difference is shown as the different magneto-optic effect. The magneto-optic effect causes changes of optical parameters of materials [12]. The optical activity of material changes with the application of magnetic field. The magneto-optic effect can give information on the change of optical characteristics of the material that can be measured through the material ability in changing the polarization angle of light wave penetrating the material. The light polarization orientation in a material caused by the external magnetic field can be learned by using Faraday rotation effect. If the fluid is used as an active material in the Faraday rotation effect, the weak external magnetic field is enough to see the rotation of polarization angle and the light intensity change. This phenomenon is interesting in developing the magnetic field sensor especially for detecting the weak magnetic field like the magnetic field within a body.

Based on the explanation above, it is paramount to study the optical characteristic of Fe_3O_4 magnetic fluid made from iron sand. In this work, specifically, the study is focused on the magneto-optic effect happening on the Fe_3O_4 magnetic fluid.

2. Experimental Method

The Fe_3O_4 magnetic fluid was prepared by using a coprecipitation- sonochemical method. The purified Fe_3O_4 powders as a main precursor was selected from natural iron sand following our previous works [13–17]. However, in this work, the synthesis method was developed by combining the coprecipitation as reported in our previous works with the sonochemical route using ultrasonic bath at a frequency of 40 KHz at room temperature for 1 hour. Furthermore, the preparation of the magnetic fluid was tracked by the previous work [18] but modified by adding more water to enhance the homogeneity of the magnetic particles. The chemical reaction of the magnetite particle formation in this work is presented in the equation (1).



The phase purity, crystal structure, and particle size of the sample were characterized by means of X-Ray Diffractometer (XRD) at ambient temperature. Furthermore, the optical properties of the magnetic fluid as a function of the external magnetic field was investigated by using a set magneto-optical experiment as presented in the following figure.

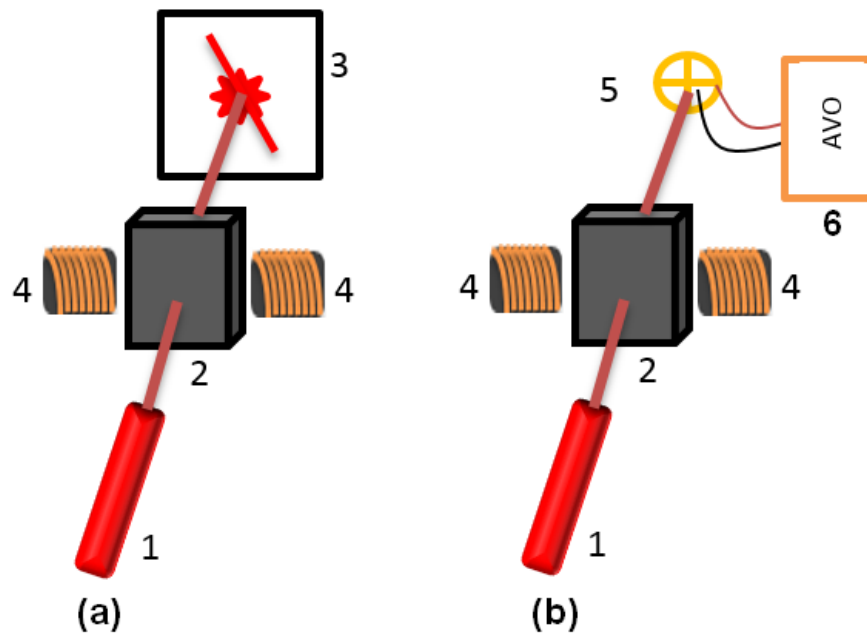


Figure 1. Experimental set-up of the optical properties of the Fe_3O_4 magnetic fluid: (a) The optical properties collected by a screen and (b) collected by optical detector

In Figure 1, number 1 represents the laser He-Ne “CVI Melles Griot” input: 115/230 VAC, max = 39.5 Hz: 50 - 400, number 2 represents the glass with the size of $2 \text{ cm} \times 1.5 \text{ cm} \times 0.3 \text{ cm}$ with the glass thickness of 1 mm, number 3 represents the screen, number 4 represents the external magnetic field, number 5 represents the optical detector “THORLABS” Si Amplified Detector, PDA 100 A – EC, number 6 represents the digital AVO-meter FLUKE 179 with the accuracy of 0.001 mV. Practically, the experiment was conducted by placing the magnetic fluid in the glass. The laser was directed to the magnetic fluid and detected by the screen and or the optical detector. The external magnetic field was varied during the experiment and measured by using a digital Teslameter MG – 3002 with the resolution of 0.01 mT. The data were collected both from the set of (a) and (b) experiments regarding Figure 1.

3. Results and Discussion

X-ray diffraction pattern of the Fe_3O_4 magnetic fluid is shown in Figure 2. Based on the quantitative analysis by using Rietica program [19], the Fe_3O_4 particle was structured in the inverse spinel with the space group of $Fd\bar{3}mZ$. The statistic parameters of the quantitative analysis were of $GoF = 0.34$, $R_{wp} = 15.18$, $R_p = 10.78$. It means that the refinement model was highly acceptable in fitting the experimental data. The lattice parameters and crystal volume were respectively $a = b = c = 8.368 \text{ \AA}$ and $V = a \times b \times c = 585.97 \text{ \AA}^3$. These results are similar to the crystal parameters of the Fe_3O_4 nanoparticles preparing from iron sand by other synthesis method [13]. Furthermore, the particle size of the Fe_3O_4 particle was approximately 9 nm after determined by using a Debye Scherer’s formula. Therefore, this particle size becomes a substantial evidence that the Fe_3O_4 particle prepared in this work is suitably dispersed in a liquid carrier to form a stable magnetic fluid.

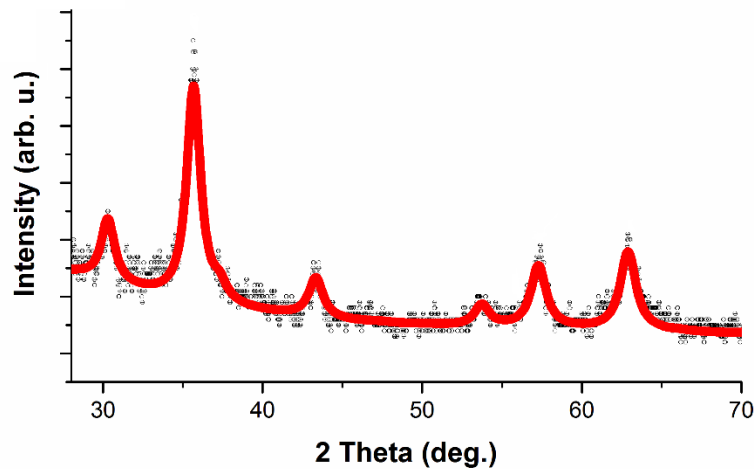


Figure 2. XRD pattern of the dried Fe_3O_4 fluid at room temperature

Figure 3 presents the effect of magnetic field on the optical characteristic of the Fe_3O_4 magnetic fluid indicated by the shift of laser beam on the screen. The amount of beam shift is shown visually in Figure 3. In Figure 3 (a), we can see that the laser light spreads over the screen. This condition happens when the laser light holds the screen directly without passing a glass beam containing ferrofluid and external magnetic field. When the glass beam containing ferrofluid is placed between laser and screen (laser shot to the glass beam containing ferrofluid), a beam arises directing to the angle of 50° as visualized in Figure 3 (b). After giving external magnetic field like in Figure (c) until Figure (n), we can see that the laser light beam seen on the screen gets widening and moving far from the x-axis.

Qualitatively, the amount of polarization angle shift influenced by the external magnetic field is shown in Figure 4. This figure shows the rotation graph of polarization angle due to the symptom of magneto-optic in the Fe_3O_4 magnetic fluid. In the range of 0 mT up to 123.8 mT, the beam shift happens in the amount of 50° to 70° . The polarization angle rotation tends to increase linearly in the magnetic field range of 0 mT to 30 mT; while above 30 mT, the polarization angle rotation increases gradually or exponentially. Generally, when a light beam penetrates an active optical object, it will transform into two light beams polarized circularly [20]. The phenomenon of polarization angle rotation happens through Faraday effect. Generally, Faraday effect is expressed as $\theta = V \times H \times I$, where V is constant of Verdet material, H is the given magnetic field, and I is the length of the optical path or the thickness of Fe_3O_4 magnetic fluid [21].

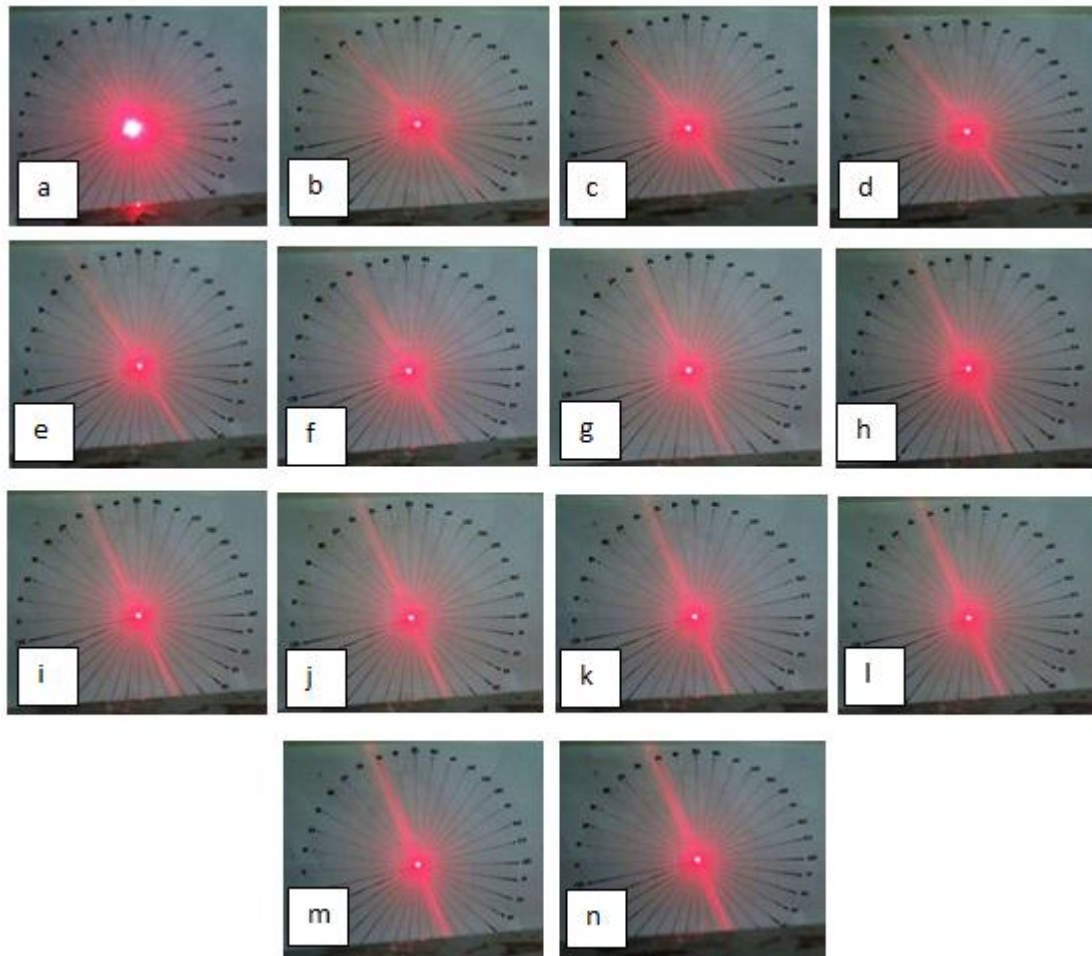


Figure 3. The polarization angle of the laser (a) for direct beam of the laser without Fe_3O_4 magnetic fluid and in the absence of external magnetic field; for the Fe_3O_4 magnetic fluid under magnetic field of (b) 0 mT (c) 0.7 mT (d) 2.2 mT (e) 3.6 mT (f) 5.1 mT (g) 21.8 mT (h) 36.4 mT (i) 50.9 mT (j) 65.5 mT (k) 80.1 mT (l) 94.7 mT (m) 109.2 mT, and (n) 123.8 mT

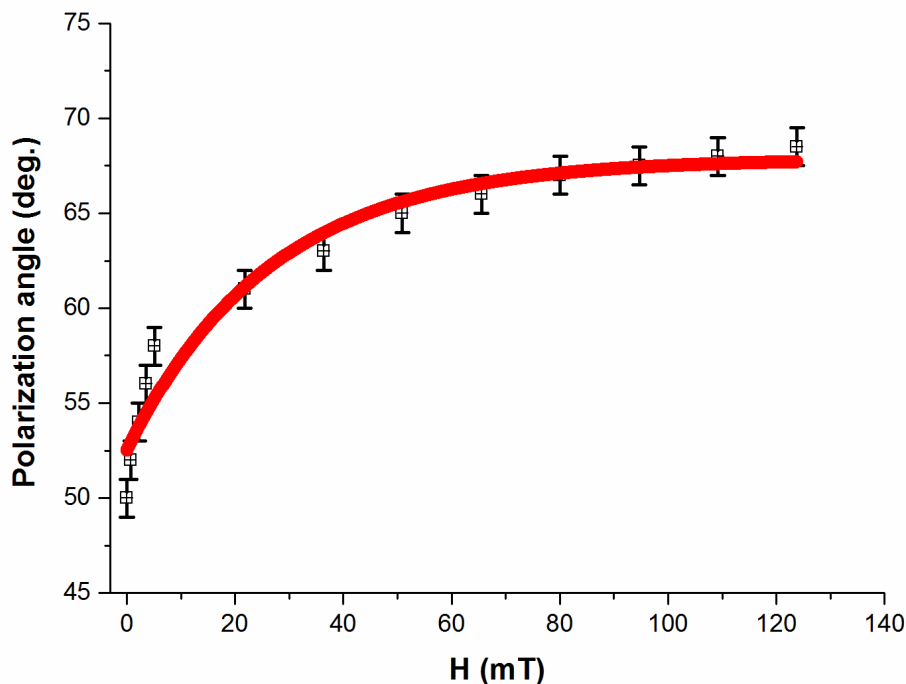


Figure 4. Polarization angle vs. external magnetic field (H) of the Fe_3O_4 magnetic fluid

Physically, the polarization angle rotation causes the increase of light intensity since the magnetic field interaction of the light wave with the external static magnetic field given to Fe_3O_4 magnetic field as a magneto-optic effect. The graph of intensity change as a function of the magnetic field is shown in Figure 5. One of the interesting phenomena in this work is the appealing of polarization although the external magnetic field was not given to the Fe_3O_4 magnetic fluid. In physics, this phenomenon occurs because laser brings the magnetic field causing the magnetic moment of Fe_3O_4 in fluid undergoes orientation. The magnetic moment orientation of Fe_3O_4 induces its particle orientation as well. Since Fe_3O_4 magnetic fluid is super-paramagnetic, how small the external field is given, it can induce the magnetic moment of the particle. Meanwhile, the flexibility of fluid compared to film, powder or bulk contribute as well to the easiness of magnetic particle orienting in the magnetic fluid. This case is different from the laser light intensity increasing linearly as a function of external magnetic field. The linear relationship between intensity and external magnetic field has an ideal potential to optical based magnetic field sensor. This experiment result is in line with the experiment result conducted by Nair et al. [14].

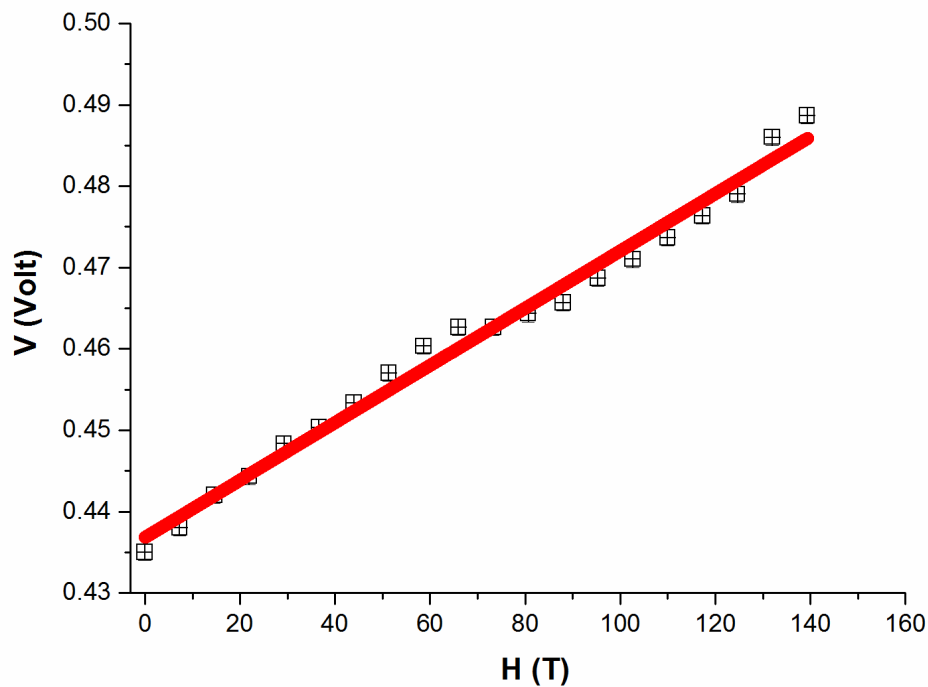


Figure 5. Voltage vs. external magnetic field of the magnetic fluids. V represents the intensity measured by using photodetector (in Volt unit)

Some reports in the literature show the occurrence of magneto-optic effect in the magnetic fluid [22,23]. The most important magneto-optic effect shown by the magnetic fluid is Faraday rotation, Faraday Ellipticity, Kerr Effect, linear dichroism, birefringence, and so forth [24]. Furthermore, it has been shown as well that Faraday rotation can happen in a railroad like-way. This quantization is correlated to the magnetic moment tunneling of resonance in the case of smaller size-quantum particles [25]. The theory of no-reciprocity demonstrates that the rotation direction of the polarized light depends on only the magnetic field direction [26]. Furthermore, the reciprocity of Faraday effect shown by the magnetic fluid and magneto-optic glass explains that although the magnetic field direction is reversed, the rotation of the polarized light is still similar to the first case [27]. Regarding this case, there are many tools developed based on the magnetic fluid such as a sensor, isolator, and modulator designed efficiently by controlling the magneto-optic effect [28,29]. Thereby, based on the result of this study, the Fe_3O_4 magnetic fluid as the preparation result of iron sand has an ideal potential to be developed further as a high sensitive optics-based magnetic sensor.

4. Conclusion

We have successfully prepared the Fe_3O_4 magnetic fluid from natural iron sand. The Fe_3O_4 magnetic fluid exhibited a magneto-optical phenomenon. The Fe_3O_4 magnetic fluid induced the change of polarization angle and its intensity originated from the external magnetic field. The change of polarization angle tended to increase exponentially as a function of external magnetic field. Furthermore, the intensity of the laser increases linearly with increasing external magnetic field. These phenomena become an essential data that the prepared Fe_3O_4 magnetic fluid opening the high potential for developing sensor, especially as a high sensitive optics-based magnetic sensor.

5. References

- [1] Zheng Y, Dong X, Chan C C, Shum P P and Su H 2015 Optical fiber magnetic field sensor based on magnetic fluid and microfiber mode interferometer *Opt. Commun.* **336** 5–8
- [2] Ke H, Huang W and Wang X 2016 Controlling lubricant migration using ferrofluids *Tribol. Int.* **93, Part A** 318–23
- [3] Sheikh L, Vohra R, Verma A K and Nayar S 2015 Biomimetically Synthesized Aqueous Ferrofluids Having Antibacterial and Anticancer Properties *Mater. Sci. Appl.* **6** 242–50
- [4] Casula M F, Corrias A, Arosio P, Lascialfari A, Sen T, Floris P and Bruce I J 2011 Design of water-based ferrofluids as contrast agents for magnetic resonance imaging *J. Colloid Interface Sci.* **357** 50–5
- [5] Rosensweig R E 2014 *Ferrohydrodynamics* (Mineola, New York: Dover Publications, Inc)
- [6] Mendelev V S and Ivanov A O 2004 Ferrofluid aggregation in chains under the influence of a magnetic field *Phys. Rev. E* **70**
- [7] Zubarev A Y and Iskakova L Y 2006 Direct and inverse domain structures in ferrofluids *Phys. Stat. Mech. Its Appl.* **367** 55–68
- [8] Laskar J M, Philip J and Raj B 2009 Experimental evidence for reversible zippering of chains in magnetic nanofluids under external magnetic fields *Phys. Rev. E* **80**
- [9] Born M, Wolf E and Hecht E 2000 *Principles of Optics: Electromagnetic Theory of Propagation, Interference and Diffraction of Light* *Phys. Today* **53** 77–8
- [10] Hopster H and Oepen H P 2005 *Magnetic microscopy of nanostructures* (Berlin; New York: Springer)
- [11] Iizuka K 2002 *Elements of photonics* (New York, N.Y: Wiley-Interscience)
- [12] Marand Z R, Farimani M H R and Shahtahmasebi N 2014 Study of magnetic and structural and optical properties of Zn doped Fe₃O₄ nanoparticles synthesized by co-precipitation method for biomedical application *Akush. Ginekol. (Sofia)* **15** 238–47
- [13] Taufiq A, Sunaryono, Rachman Putra E G, Okazawa A, Watanabe I, Kojima N, Pratapa S and Darminto 2015 Nanoscale Clustering and Magnetic Properties of Mn_xFe_{3-x}O₄ Particles Prepared from Natural Magnetite *J. Supercond. Nov. Magn.* **28** 2855–63
- [14] Pratapa S, Susanti L, Insany Y A S, Alfati Z, Hartono B, Mashuri, Taufiq A, Fuad A, Triwikantoro, Baqiya M A, Purwaningsih S, Yahya E and Darminto 2010 XRD line-broadening characteristics of M-oxides (M = Mg, Mg-Al, Y, Fe) nanoparticles produced by coprecipitation method *AIP Conf. Proc.* **1284** 125–8
- [15] Sunaryono, Taufiq A, Munaji, Indarto B, Triwikantoro, Zainuri M and Darminto 2013 Magneto-elasticity in hydrogels containing Fe₃O₄ nanoparticles and their potential applications *AIP Conf. Proc.* **1555** 53–6
- [16] Sunaryono, Taufiq A, Putra E G R, Okazawa A, Watanabe I, Kojima N, Rugmai S, Soontaranon S, Zainuri M, Triwikantoro, Pratapa S and Darminto 2015 Small-Angle X-Ray Scattering Study on PVA/Fe₃O₄ Magnetic Hydrogels *Nhano* **11** 1650027
- [17] Sunaryono, Taufiq A, Mashuri, Pratapa S, Zainuri M, Triwikantoro and Darminto 2015 Various Magnetic Properties of Magnetite Nanoparticles Syntesized from Iron-Sands by Coprecipitation Method at Room Temperature *Mater. Sci. Forum* **827** 229–34
- [18] Taufiq A, Sunaryono, Rachman Putra E G, Pratapa S and Darminto 2015 Nano-Structural Studies on Fe₃O₄ Particles Dispersing in a Magnetic Fluid Using X-Ray Diffractometry and Small-Angle Neutron Scattering *Mater. Sci. Forum* **827** 213–8
- [19] Hunter B A 1998 Rietica - A visual Rietveld program *Newsl. Int. Union Crystallogr. Comm. Powder Diffraction*
- [20] Khandanlou R, Ahmad M, Shameli K and Kalantari K 2013 Synthesis and Characterization of Rice Straw/Fe₃O₄ Nanocomposites by a Quick Precipitation Method *Molecules* **18** 6597–607
- [21] Nair S S, Rajesh S, Abraham V S and Anantharaman M R 2010 Ferrofluid Thin Films as Optical Gaussmeters Proposed for Field and Magnetic Moment Sensing *Indian Acad. Sci.* **34** no 2 245–9

- [22] Lacoste D, Donatini F, Neveu S, Serughetti J A and Van Tiggelen B A 2000 Photonic Hall effect in ferrofluids: Theory and experiments *Phys. Rev. E* **62** 3934–43
- [23] Davies H W and Llewellyn J P 1980 Magneto-optic effects in ferrofluids *J. Phys. Appl. Phys.* **13** 2327–36
- [24] Kooij E S, Gâlcă A C and Poelsema B 2006 Versatile transmission ellipsometry to study linear ferrofluid magneto-optics *J. Colloid Interface Sci.* **304** 261–70
- [25] Jamon D, Donatini F, Siblini A, Royer F, Perzynski R, Cabuil V and Neveu S 2009 Experimental investigation on the magneto-optic effects of ferrofluids via dynamic measurements *J. Magn. Magn. Mater.* **321** 1148–54
- [26] Donatini F, Jamon D, Monin J and Neveu S 1999 Experimental investigation of longitudinal magneto-optic effects in four ferrite ferrofluids in visible-near infrared spectrum *IEEE Trans. Magn.* **35** 4311–7
- [27] Deb P, Gogoi M and Karmakar P K 2012 Anomalous magneto-optic Faraday rotation behavior due to resonant tunneling of magnetic moment *J. Opt.* **41** 41–7
- [28] Massard C, Taverdet J L, Brouillet S and Donnet C 2007 Hybrid Sol–gel thin films for magneto-optical applications: Chemical, optical and tribological study *Surf. Coat. Technol.* **202** 1067–72
- [29] Wang S, Sun C, Du L, Yao C and Yang Y 2012 Reciprocity of Faraday effect in ferrofluid: Comparison with magneto-optical glass *Opt. - Int. J. Light Electron Opt.* **123** 553–8

Acknowledgements

The authors (AT and AH) very appreciate DRPM-KEMENRISTEKDIKTI, Republic of Indonesia, for supporting “HIBAH PENELITIAN” 2016-2017.