Investigation of Effective Permeability and Dielectric aspect of Dilute Magnetic Dielectrics by Optical Method

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The objective of this paper is to investigate the magnetic nature of Dilute Magnetic Dielectrics (DMD) under variation of external magnetic field. The said variation is studied over developed nano sized Gadolinium Nickel Sulfide complex, Cobalt Sulfide, Nickel Sulfide and Titanium Sulfide as a DMD system. The observed experimental field variation of the effective magnetic permeability is the analyzed results of optical experiment. The experiment records the variation of Brewster angle of incident polarized LASER beam from the surface of developed DMD specimen with applied out of plane external magnetic field. The relative permittivity and relative magnetic permeability were estimated by following the electromagnetic theory. The overall results obtained are found to be holding a good agreement between theory and experiment.

Introduction

The importance of composite dielectric materials in present day technology has been emphasized in many literatures [**1**]. In this work a simple form of dielectric behavior of dilute magnetic dielectric (DMD) type composite material is formulated. In an early work [**2**] the effective dielectric response of the system under external magnetic field was studied theoretically. The work provides a good account of effective dielectric response of the DMD system.

Long range magnetic order is mainly found in materials containing elements from the 3d transition metals (such as Co and Ni) and the 4f rare-earth metal series like Gd. Magnetic materials which contain 4f electrons usually have a saturation magnetization close to what has been predicted by Hund's rule, but the moment configuration is quite complicated and spin reorientations are often observed. In this work authors have investigated dielectric response for four DMD sample i.e., Gadolinium Nickel Sulfide Complex, Cobalt Sulfide, Nickel Sulfide and Titanium Sulfide. All samples were synthesized following chemical and green techniques. Later process provides good stability of the nano clusters (NC) due to in-situ capping of sample NC. It has been found that the optical band gap in sample developed by green synthesis is lowered considerably over that in the chemically synthesized sample [**3**]. The green agencies used in this work are Jatropha latex [**4**] and dilute Garlic extracts, both are enriched in sulphur and other non-ferric molecules. Jatropha latex has some ethno medical uses like wound healing, blood coagulation activities.

Particularly, the use of latex, proteins and phyto chemicals for the synthesis of metal nano particles has dual advantage that they not only act as a reducing agent but also act as a capping agent and deter the particle aggregation [**4**].

One of the most popular techniques for detection and measurement of magnetism is the optical method. The magneto-optic Kerr Effect (MOKE) [**5**] is a powerful spectroscopic tool in magnetic materials research. Magneto-optic phenomena play an important role in the development of Maxwell's electro-magnetic (EM) theory. Maxwell theory describes a macroscopicdescription of the MOKE involving the energy and material dependent dielectric tensor and the magnetic aspects. The microscopic manifestation of the magneto-optic effect lies in the interaction between the electric field of the incident EM wave and the magnetic moments in the solid. Magneto Optical Kerr effects are generally described macroscopically by dielectric tensor theory or the effects can also be described microscopically, where the coupling between the electric field of the light and the magnetisation occurs by the spin-orbit interaction. To understand the magneto optical Kerr effects, one need to understand terminologies associated with the effect, how the state of polarization of reflected light is dependent upon the initial polarization and the magneto optical geometry in which it is being used. The MOKE instrumentation is an extremely elite technology.

In this present work another simple optical techniques for investigation of magnetism in a DMD is employed. The technique exploited the Maxwell's EM theory. DMD with good reflecting surface to exhibit minimum reflectance of incident in plane polarized light. The corresponding angle of incidence is the Brewster angle [**5**]. Brewster's no reflection condition is one of the main features of the laws of reflection and refraction of electromagnetic waves at interface between two media. For a specific incident angle, known as the Brewster angle, the reflected wave vanishes. In dielectric media, this phenomenon exists only for transverse-magnetic (TM) waves (p waves), and not for transverse-electric (TE)waves (s waves). The influence of external out of plane magnetic field is to cause the variation of the Brewster angle due change in surface magnetism of the specimen. In this present work the experimental setup involves a monochromatic light source which is relativized by a stable laser diode, the latter provides a monochromatic, nearly parallel light beam of roughly linearly polarized light. Further elements are a polarizer P, the magnetic sample S, an analyzer A, and the photo detector. According to Electro Magnetic Theory, light that is reflected from a magnetized surface suffer change in both polarization and reflected intensity. The effect is similar to the Faraday-effect. The Faraday-effect describes changes to light transmitted through a magnetic material, while the Kerr effect [**6**] describes changes to light reflected from a magnetic surface. Both effects result from the off-diagonal components of the dielectric tensor ε. These off-diagonal components give the magneto-optic material an anisotropic permittivity, meaning that its permittivity is different in different directions.

Theory

The permittivity affects the speed of light in a material:

$$V= (\mu\varepsilon)^{-1/2} \tag{1}$$

Physically, Brewster's phenomena can be understood as follows, when electromagnetic waves is at the interface between two media, the direction of the induced electric dipole in second medium is perpendicular to the wave vector. With regard to TM waves, the dipole lies in the plane of incidence. A linearly vibrating dipole radiates transversally and cannot emit radiation in the direction of the vibration. This direction coincides with the wave vector of the reflected wave when the Brewster condition is satisfied. The oscillating dipoles in the second medium do not send any waves in the direction of the reflection. On the other hand, with regard to TE waves, each dipole is perpendicular to the plane of incidence and emits wave isotropically in the plane. Therefore, no special angles exist for TE waves. The dipole model also explains the sign change in the amplitude reflectivity when the angle is changed through the Brewster angle.

According to Electromagnetic theory, at the interface of two dielectrics,

$$E_{0I}-E_{0R}=\beta E_{0T} \tag{2}$$

Where, E_{0I}, E_{0R} and E_{0T} are the amplitudes of incident, reflected and transmitted electric field of EM wave respectively.

$$\beta=\mu_1 v_1/\mu_2 v_2 \tag{3}$$

For Non-magnetic dielectric

$$\mu_1 = \mu_2 = \mu_0 \tag{4}$$

Brewster angle (θ_B),

$$\theta_B = \tan^{-1}\beta = \tan^{-(n2/n1)} \tag{5}$$

Where, n2 and n1 are absolute refractive indices of the two medium.

For DMD system, in general

$$\tan\theta_B = (\mu_r\varepsilon_r)^{-1/2} \tag{6}$$

At zero field the relation given by equation (6) becomes,

$$\tan\theta_B(0) = (\mu_r(0)\varepsilon_r)^{-1/2} \tag{7}$$

And at the presence of field H. the same is,

$$\tan\theta_B(H) = (\mu_r(H)\varepsilon_r)^{-1/2} \tag{8}$$

Hence using equations (7) and (8),

$$\mu_r(H)/\mu_r(0) = [n(H)/n(0)]^2 \tag{9}$$

Ferromagnetic or super-paramagnetic [**6**] material exhibits a non-linear magnetization curve and hence μ_r (H) is a non-linear function of H. Moreover, as H→ H_K, the relative permittivity is $\mu_r \rightarrow 0$, where H_K is saturation field.

The values of Re and Im part ε (ω) may be estimated directly from Dielectric Spectroscopy data.

Experimental

Materials and material synthesis

Latex of Jatropha curcas L. was collected early in the morning because production of latex is in higher amount during early morning. Crude latex was obtained by cutting the green stems of J. curcas plants. Milky white latex was stored at -40^0 C until further use. All the aqueous solutions were prepared in triple-distilled de-ionized water. In typical reaction mixture, 1 mL crude latex was diluted to 300 mL using triple-distilled deionized water to make it 0.3 %, and 20 mL of this latex solution was mixed with 20 mL 2.5 mM Gadolinium (III) acetate hydrate, analytical grade (Alfa Aesar) aqueous solution. Now the mixture was allowed to stand at room temperature (27^0C) in laboratory ambience with continuous stirring with the help of magnetic stirrer for 24-48 h. After 25 h, solution was observed to have distinctly makeable yellowish-brown clusters deposited at the bottom of the flask. After the completion of reaction, the precipitate was washed several times with distilled water. The final precipitate was dried at 50^0C for 1 h in a vacuum oven. Gadolinium Sulfide sample were prepared following the process of preparation of ZnS [**4**].

Gadolinium Nickel Sulfide complex sample were prepared by solid-state reaction method using high purity precursor materials of Gadolinium Sulfide and NiO. Stoichiometric proportion of the powders were thoroughly mixed and heated at temperatures about 50^0C for 1 h. The Cobalt Sulfide, Nickel Sulfide and Titanium Sulfide are prepared by the same process with latex of J. curcas L of respective acetate hydrate.

The mentioned process ensure formation sulphate compound of the metallic element. In practice formation metallic sulphate from corresponding sulphide always requires a temperature > 300^0c. Pellet of developed samples was formed by mechanical pressing at pressure 12 ton/cm^2. This applied pressure makes the specimen shiny enough to achieve optical reflection.

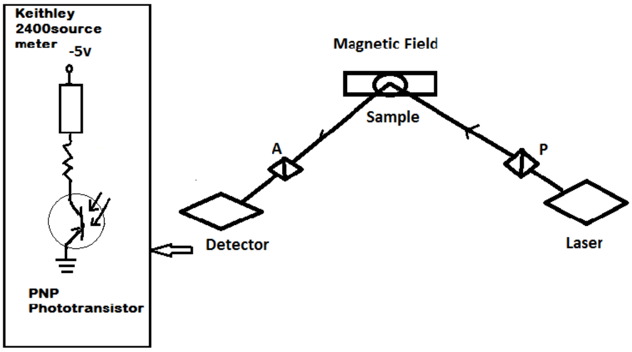

Fig. 1. An indigenous experimental setup.

Response measurements

The optical reflectance measurement was carried out using specially designed optical spectrometer, an indigenous set up in which a monochromatic He-Ne LASER (wave length 633 nm) was used as a source and a polarizer was used at the collimator end of the spectrometer to get plane polarized light. The cross-wire of the spectrometer was replaced by a photo transistor to produce equivalent current proportional to the light intensity. The electric current equivalent of the reflected light intensity for different angle of incidence (at angular resolution of 20") of the plane polarized light was recorded by photo transistor based high sensitive detector along with Keithley 2400 (USA) source meter. Experiment was carried out for in plane polarized light namely polarization parallel to the plane of incidence. All measurements were done at room temperature, 300K. The developed pellet was sandwiched between two high polished copper plates to form experimental parallel plate capacitor with known geometry. The measurement was done at room temperature (RT) with HIOKI 3522-50 LCR Hi TESTER (JAPAN). DC current voltage characteristics of Gadolinium Nickel Sulfide complex, Cobalt Sulfide, Nickel Sulfide and Titanium Sulfide were measured by Keithley 2400 (USA) source meter.

Results and discussion

The optical reflectance measurements using the mentioned set were carried over the developed DMD specimens at different external fields.

Fig. 2 Shows the variation of μr(H)/μr(0) with H. The overall variation nature is that of Ferro-magnetic/ superparamagnetic [7] material. A knowledge of μr(0) or εr may be used in evaluation of μr(H). The same may be applied by neglecting small contribution from cross coupling effect. The results of variation of μr(H)/μr(0) with H, shows the typical character of ferro-magnetic /superparamagnetic material. **Fig. 2(a)** exhibits a very superparamagnetic clear signature of ferro-magnetic/ superparamagnetic material which may be obtained from derivative of magnetization curve. The graph also shows the tendency to assume the magnetic saturation of the material at low field. This nature is also observed in **Fig. 2(b)** and **Fig. 2(c)** but saturation at relatively higher field compared to that in **Fig. 2(a)**. The material corresponding to **Fig. 2(d)** exhibits a typical nature for ordinary paramagnetic material.

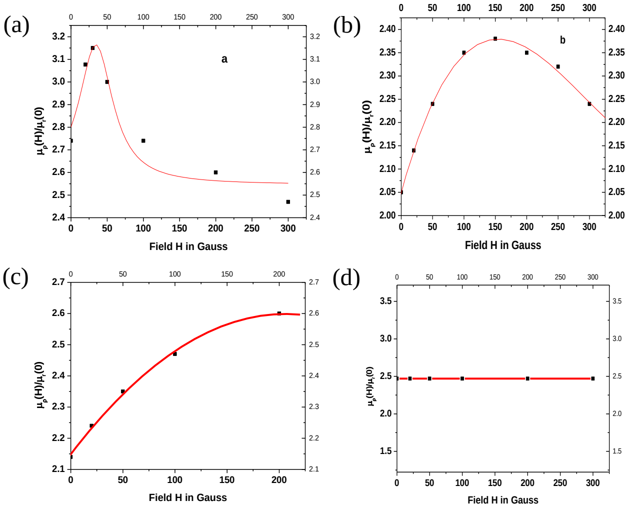

Fig. 2. Plot of variation of $\mu_r(H)/\mu_r(0)$ of the specimen with external field (a) Gadolinium Nickel Sulfide complex(1.51mm), (b) Cobalt Sulfide (1.54mm), (c) Nickel Sulfide (1.56mm) and (d) Titanium Sulfide (1.52mm). Dots are the experimental points and solid red line is best fit curve for the relation.

They are relatively good candidates for DMD system. The experimental analysis indicates that obtained magnetism is mostly due to the surface magnetism [**8**] which is very interesting for topological crystalline insulators [**9**].

Following Stoner criterio and beyond it may be emphasized that surface magnetism is supposed to be more pronounced over Bulk counterpart due to enhanced moment at the surface. Surface magnetism of nano-structured material exhibits many interesting features [**10**]. **Fig. 3(a,b,c)** show the variation of relative ε with frequency of impressed a.c. signal. They exhibit a strong dielectric nature of materials. The extrapolated dc vale of ε decreases for material corresponding to **Fig. 3(a)** to **Fig. 3(d)**.

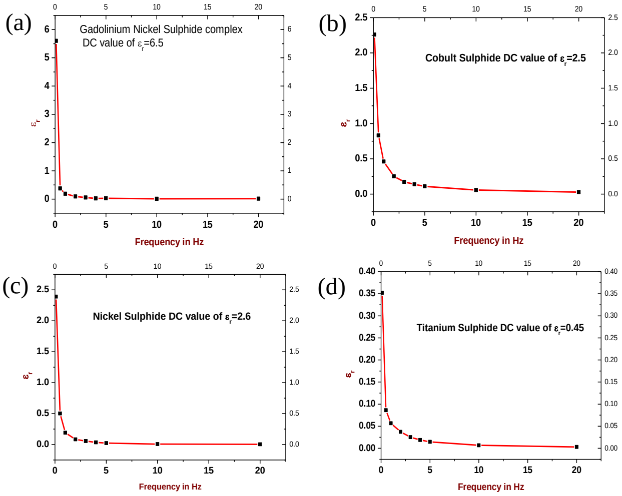

Fig. 3. Plot of variation of relative ε with frequency of impressed a.c. signal. (a) Gadolinium Nickel Sulfide complex, (b) Cobalt Sulfide, (c) Nickel Sulfide and (d) Titanium Sulfide.

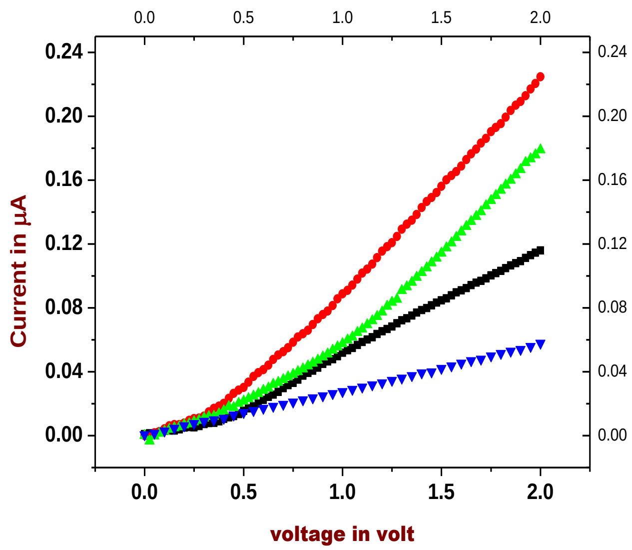

Fig. 4. Plot of variation of dc current voltage characteristics of (a) Gadolinium Nickel Sulfide complex(black), (b) Cobalt Sulfide (red), (c) Nickel Sulfide (green) and (d) Titanium Sulfide (blue).

Fig. 4 shows the dc volt-ampere characteristics of Gadolinium Nickel Sulfide complex, Cobalt Sulfide, Nickel Sulfide and Titanium Sulfide. Computed value of dc conductivity from linear part of respective curves in **Fig. 4** shown in **Table 1** and it shows that Titanium Sulfide has lowest electrical conductivity hence poorest among the developed DMD.

Table 1. Computed value of dc conductivity unit=µ S/cm from linear part of respective curves from Fig.4.

dc conductivity unit=µ S/cm			
Cobalt Sulfide	Nickel Sulfide	Gadolinium Nickel Sulfide complex	Titanium Sulfide
5.88211E-4	3.19E-4	2.87064E-4	1.06E-4

Conclusion

The variation of Brewster angle with external field on the developed DMD was recorded successfully. The experimental DMD has a Ferro-magnetic or super-paramagnetic nature at room temperature, an improved version of the instrumentation could be a cost effective one for determination magnetism in such DMD specimens. The overall success of this investigation is found to be good and concise.

Conflicts of interest

There are no conflicts to declare.

Supporting information

Supporting informations are available onlineat journal website.

Keywords

Magnetoelectric effect, super paramagnetism, surface magnetism.

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