

Band structure of a 2D photonic crystal based on ferrofluids of $\text{Co}_{0.8}\text{Zn}_{0.2}\text{Fe}_2\text{O}_4$ nanoparticles under perpendicular applied magnetic fields

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Abstract. Using a ferrofluid of cobalt-zinc ferrite nanoparticles ($\text{Co}_{0.8}\text{Zn}_{0.2}\text{Fe}_2\text{O}_4$) coated with oleic acid and suspended in ethanol, we have fabricated a 2D photonic crystal (PC) by the application of an external magnetic field perpendicular to the plane of the ferrofluid. The 2D PC is made by rods of nanoparticles organized in a hexagonal structure. By means of the plane-wave expansion method, we study its photonic band structure (PBS) which depends on the effective permittivity and on the area ratio of the liquid phase. Additionally, taking into account the Maxwell-Garnett theory we calculated the effective permittivity of the rods. We have found that the effective refractive index of the ferrofluid increases with its magnetization. Using these results we calculate the band structure of the photonic crystal at different applied magnetic fields, finding that the increase of the applied magnetic field shifts the band structure to lower frequencies with the appearance of more band gaps.

1. Introduction

Magnetic nanofluids or ferrofluids are part of a new class of magnetic materials. They exhibit magnetic and fluid properties simultaneously, becoming materials of technological importance [1]. Magnetic fluids have attracted a great deal of attention of scientist and engineers, not only because their structural patterns under external magnetic fields where periodic columns are formed in the ferrofluid by a applied perpendicular magnetic field [2], but also by its optical properties, such as birefringence, optical negative refraction, diffraction, light transmission and many potentially electro-optical characteristics of the magnetic fluids, for example, the optical switch, the tunable grating, and photonic crystals [3, 4, 5].

2. Experimental method

The size and physical properties of the nanoparticles depend on preparation parameters such as, reaction temperature, pH of the suspension, initial molar concentration, and others. In this case, the $\text{Co}_{1-x}\text{Zn}_x\text{Fe}_2\text{O}_4$ magnetic nanoparticles were prepared using the chemical co-precipitation method, which is probably the most-common and the most-used synthesis of ferrofluids or magnetic fluids based on magnetic nanoparticles. The preparation of surfacted and ionic aqueous ferrofluids based on spinel ferrite nanoparticles has been reported by Massart [6]. The preparation of our ferrofluid samples are described in the previous work of J. Lopez et



al. [7]. In their work they describe a complete process to the synthesis of $\text{Co}_{1-x}\text{Zn}_x\text{Fe}_2\text{O}_4$ with nominal values of $x = 0.25, 0.50$ and 0.75 .

3. Theoretical Framework

For the transversal magnetic mode (TM), the wave equation of the magnetic field $\mathbf{E}_{\mathbf{k}}$ may be obtained from Maxwell's equations and expressed as

$$\frac{1}{\epsilon(\mathbf{r})} \nabla \times (\nabla \times \mathbf{E}_{\mathbf{k}}) = \frac{\omega^2}{c^2} \mathbf{E}_{\mathbf{k}}, \quad (1)$$

where k represents the wave propagation vector of the mode and $\epsilon(r)$ is the position dependent dielectric constant of the periodic structure. From Bloch theorem and theorems of Fourier analysis, the electromagnetic fields in a periodic medium can be written as

$$\mathbf{E}_{\mathbf{k}}(\mathbf{r}) = \sum_{\mathbf{G}} \mathbf{E}_{\mathbf{k}}(\mathbf{G}) e^{i(\mathbf{k}+\mathbf{G})\mathbf{r}}, \quad (2)$$

where $\mathbf{G}$ represents a lattice vector in reciprocal space, describing the periodic structure, similarly a periodic dielectric function can be expanded as

$$\frac{1}{\epsilon(\mathbf{r})} = \sum_{\mathbf{G}} \chi(\mathbf{G}) e^{i\mathbf{G}\cdot\mathbf{r}}, \quad (3)$$

where $\chi(\mathbf{G})$ are the Fourier expansion coefficients [8].

By substituting (2) and (3) into (1) a matrix eigenvalue problem is obtained, where for a fixed wave vector $\mathbf{k}$, the frequency ω of the allowed modes in the periodic structure are found as eigenvalues.

To characterize electromagnetic properties of composite media, we use the Maxwell-Garnett approximation (MGA). The following equation is the exact solution for the equivalent permittivity of the core-shell composite nanoparticle [9]

$$\frac{\epsilon_r - \epsilon_a}{\epsilon_r + 2\epsilon_a} = p \frac{\epsilon_F - \epsilon_a}{\epsilon_1 + 2\epsilon_a}, \quad (4)$$

where $p = \frac{r^3}{(r+d)^3}$ is the volume ratio of the nanoshell to the whole coated nanoparticle, r is the radius of the core ($\text{Co}_{0.8}\text{Zn}_{0.2}\text{Fe}_2\text{O}_4$) with permittivity ϵ_F , and d is the thickness of the shell (oleic acid) with permittivity ϵ_a .

4. Results and discussion

In order to consider the effects of the applied magnetic field on the PBS, we have used the data given in the work by Ramana [10], where it is established the dependence of the dielectric constant of the nanoparticles with the Zn concentration. In our numerical calculations, we have taken ϵ_F to be 18 for $\text{Co}_{0.8}\text{Zn}_{0.2}\text{Fe}_2\text{O}_4$ ferrite, $\epsilon_a = 2.34$, and $r = 4.8\text{nm}$ and $d = 5.8\text{nm}$ [7].

Since the structural pattern is very important for the calculation of the PBS and varies significantly with the perpendicular magnetic field, in Fig.1 we present the structural evolution of the magnetic ferrofluid film with a magnetic concentration of 0.16 emu/g , injected into a glass cell with an area of 18mm^2 and a depth ranging from 10 microns, and under three values of the applied magnetic field: 0, 73 and 217 Oe. From Fig.1 it is observed that with the increasing of field strength, more magnetic columns are formed and the area occupied by the liquid phase is reduced.

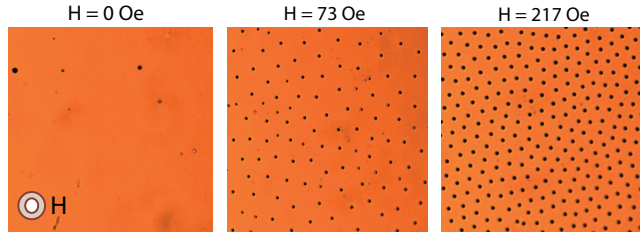


Figure 1. (Color online) Optical images of $\text{Co}_{0.8}\text{Zn}_{0.2}\text{Fe}_2\text{O}_4$ magnetic ferrofluid films under perpendicular magnetic field taken at room temperature.

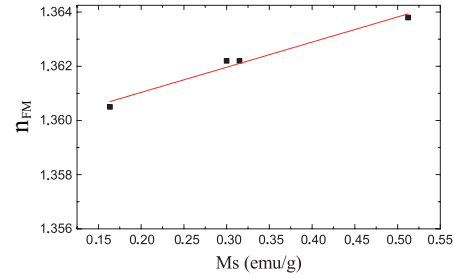


Figure 2. Linear relationship between the refractive index and the magnetic concentration of ferrofluid film under zero field.

With the values given above for ε_F , ε_a , and using Eq.5 we find $\varepsilon_r = 6.77$ for each column. For the PC structured in a hexagonal pattern, the ratio d/a of the diameter d to the period a of the magnetic columns is related to the area ratio through

$$\frac{d}{a} = \sqrt{\frac{2\sqrt{3}}{\pi} \frac{A_{col}}{A}}, \quad (5)$$

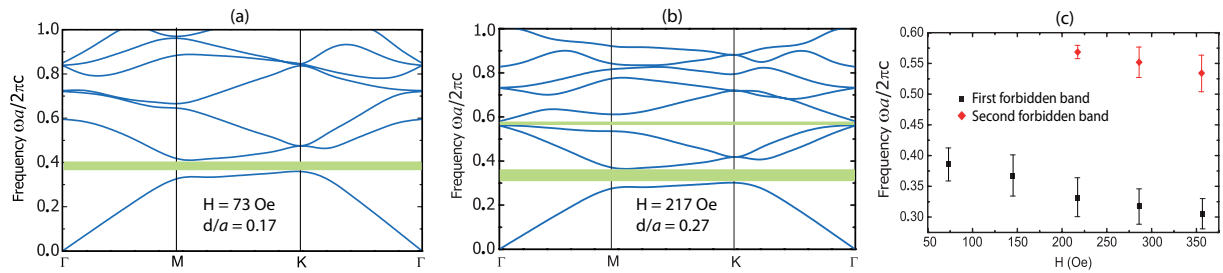


Figure 3. (Color online) Band diagrams of $\text{Co}_{0.8}\text{Zn}_{0.2}\text{Fe}_2\text{O}_4$ magnetic ferrofluid photonic crystals for TM mode at two different (a-73Oe, b-217Oe) strengths of magnetic field. c) First and second band gaps of TM mode as function of the magnetic field strength, the vertical bars mark the width of the band gaps.

where A_{col} denotes the total area of the cross sections of the columns within the area A of the magnetic fluid film. For the calculation of the dielectric constant of the liquid phase, which varies with the field strength, we found the experimental relationship between the refractive index n_{MF} and the concentration M_s of the magnetic fluid film under zero field, as it is presented in Fig.2, and given by

$$\varepsilon_{liq}(H) = n_{FM}^2(H = 0) = (0.01M_s + 1.36)^2 \quad (6)$$

To obtain the effective permittivity of the liquid phase, the refractive index of the magnetic ferrofluid with $M_s = 0.16, 0.30, 0.32$ and 0.51 emu/g , was measure under zero field. In the calculations the magnetic permeability of the ferrofluid is taken to be 1, due to that $\text{Co}_{1-x}\text{Zn}_x\text{Fe}_2\text{O}_4$ magnetic ferrofluid presents a tendency to super-paramagnetic behaviour at room temperature

Using the plane wave method, the PBS for the TM mode of PCs based on ferrofluids with hexagonal structure were calculated. The PBS at two different strengths of magnetic field are plotted in Fig.3. The frequency has been normalized to $2\pi c/a$ where c is the light speed in

vacuum. We found that for each of the directions $\Gamma M - MK - K\Gamma$ inside the PC there exist partial band gaps [8] as it is shown in Fig.3, where the radiation for a given wavelength cannot propagate inside the PC, namely, there are partial band gaps in the PC under some specific angles. Also, for this mode the number of the forbidden band gaps increases with the magnetic field strength from 1 to 2 band gaps, where the thickness of the first one does not vary in the range between 73 Oe and 217 Oe and it is shifted to lower frequencies. In addition, it is observed that the PBS is shifted to lower frequency regions. This behavior is related to the increase of the filling factor with the magnetic field strength, where more rods appear in the PC structure, causing an area reduction of the liquid phase, which should cause an increment of the dielectric constant of the material that minimize the energy, in accordance to the electromagnetic variational theorem. From the Fig. 3c we can see the existence of band gaps for different magnetic field strengths in the TM mode. This behavior is explained by the high refractive index contrast in the PC.

5. Conclusions

Ferrofluids based $\text{Co}_{0.8}\text{Zn}_{0.2}\text{Fe}_2\text{O}_4$ ferrite nanoparticles coated by oleic acid and suspended in ethanol were prepared by co-precipitation method. Ferrofluid placed in an external magnetic field shows a 2D PC made by rods of nanoparticles organized in a hexagonal structure. Using the plane-wave method, we theoretically studied the PBS which depends on the effective permittivity of the liquid phase, the area ratio of the ferrofluid, and the effective permittivity of the rods. We have found that the PBS is shifted to lower frequencies with the augment of the magnetic field. On the other hand, we found that the band gaps increase with the magnetic field strength.

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