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Direct observation of magnetically induced attenuation and enhancement of coherent backscattering of light

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Vanishing of the forward as well as the backward-scattered laser light from a dispersion of micron sized magnetic particles in a ferrofluid is observed at a certain critical magnetic field H_c. Upon increasing the field the forward-scattered intensity remains zero but the backward-scattered intensity enhances by a large amount. The transmitted intensity remains zero up to a certain field value and exhibits a stop band of field similar to a stop band of frequencies observed in other photonic materials. Results are discussed in light of existing theories. The remarkable simplicity of tuning the effects will be useful to develop photonic devices.

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I. INTRODUCTION

Evidence of localization and enhanced coherent backscattering of light has opened new vistas in research work on photonic effects and photonic materials.[1–6] Multiple scattering in strongly scattering disordered media, interference effects in partially ordered materials, and quasicrystalline materials are usually used to observe these effects.[1,7–10] In all these investigations nonmagnetic scatterers surrounded by a nonmagnetic medium are used. Busch and John[6] discussed the possibilities of tuning the light localization by applying an electric field to nematic liquid crystal. The case of nonmagnetic scatterers embedded in a Faraday active medium is experimentally studied by Lenke *et al.*[11] Effects of magnetic field on coherent backscattering of light by Faraday active Mie scatterers surrounded by nonmagnetic medium are analyzed by Lacoste and van Tiggelen[12] and by Martinez and Maynard.[13] Multiple scattering of light by disordered magnetic scatterers placed in a nonmagnetic medium is theoretically investigated by Pinheiro *et al.*[14–16] They have shown that the magnetic scattering favors the localization of light. Earlier theory for single scattering of light by a magnetic sphere was developed by Kerker *et al.*[17] They have predicted several unusual effects. When the relative dielectric constant ε and the relative magnetic permeability μ of the magnetic scatterers are equal, it is shown that backscattering will be zero and these is no depolarization. In case of Rayleigh scatterers it is shown that when $\varepsilon=(4-\mu)/(2\mu+1)$ the forward-scattered intensity becomes zero. Both the conditions are difficult to satisfy under normal conditions. Recently the Mie scattering intensity of a magnetic sphere has been derived by Tarento *et al.*[18] They have extended the classical Mie scattering problem to a media where the dielectric constant ε is a tensor with a gyrotropic term. They have shown that for small magnetic spheres magnetism affects the Mie scattering. It may be noted that in both the theories the surrounding medium is considered to be nonmagnetic. Experimental verification of the above unusual scattering effects was not so far available. Recently, experimental evidence of zero forward scattering by micron sized magnetic spheres dispersed in a ferrofluid was given by Mehta *et al.*[19] It was shown that at a certain critical field H_c the diffraction patterns in the forward direction disappear. The field H_c was found to depend on the concentration of the ferrofluid as well as on the size of the magnetic spheres. Considering the fact that both the dielectric constant (ε_f) and permeability (μ_f) of the ferrofluid can be varied by applying an external magnetic field H it was argued that the ε and μ of the micron sized magnetic spheres with respect to the ferrofluid will also vary with applied field. Both (ε_f) and (μ_f) also depend on the direction of the applied magnetic field and (ε_f) depends upon the direction of the electric vector of the incident light.[20,21] At a certain field H_c the condition for zero forward scattering may be satisfied. If we assume that Rayleigh-Gans theory is applicable for the micron sized particles then the observed effect can be explained on the basis of Kerker's theory.[19] Other photonic effects in the dispersion were also found and are described in this paper. We compare the magnetically induced forward as well as backward-scattering patterns of light. It is observed that at a certain critical field H_c both the patterns disappear. A slight increase in the field enhances the backscattered intensity significantly while the forward-scattered intensity remains zero up to certain field, i.e., it exhibits a distinct stop band of field. This stop band is affected by the addition of a small amount of nonmagnetic impurities. Thus in such dispersions we have a magnetically tunable photonic band gap (MPBG) similar to that found optically tunable band gap in other photonic materials.[6,22]

The paper is organized as follows: Section II deals with a brief description of the samples and the experimental setup. In Sec. III results of the investigations are described. Results are analyzed in Sec. IV. Summary of the present findings is given in Sec. V.

II. EXPERIMENTAL

Three different materials are used in the investigations viz. (a) ferrofluid, (b) magnetic scatterers, and (c) nonmagnetic scatterers.

A. Ferrofluid

A ferrofluid,[23] also known as magnetic fluid, is a colloidal dispersion of single domain or subdomain sized ferro or ferrimagnetic particles in a liquid matrix. Depending upon the type of applications or investigation one can select the host

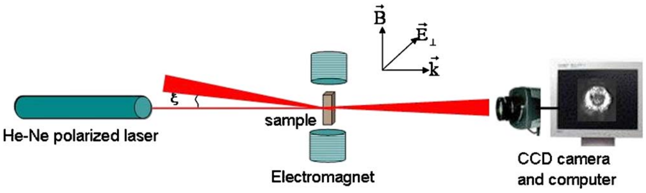

FIG. 1. (Color online) Schematic of the experimental setup.

liquid as well as the magnetic material. Colloidal stability is maintained either by steric repulsion or by Coulomb repulsion.[20] When number density of the dispersed magnetic particles is large ($\approx 10^{23}$) the fluid exhibits several novel hydrodynamic phenomena under the influence of externally applied magnetic field.[23] Their physical properties like viscosity, density, refractive index, etc., are modified by the applied field. Though the medium is not strictly homogeneous but considering the nanometer size of the particles continuum mechanics is applied to account for several magnetically induced effects in such fluids.[23]

In the present investigations nanomagnetic particles of magnetite were synthesized by a coprecipitation technique.[24] The particles were coated by oleic acid and dispersed in kerosene. The fluid was found to be stable and no aggregates were detected after centrifugation at 12 000 rpm. The saturation magnetization (M_s) of the fluid was 200 Gauss and the domain magnetization of the magnetic particles was 320 emu/cc. Assuming lognormal distribution of sizes the average diameter of the particles found from the magnetization study was 10 nm with the standard deviation in logarithmic of particle diameter $\sigma=0.3$ (Refs. 25 and 26). This fluid was diluted as per the experimental requirement.

B. Suspension of micron sized magnetite particles

Commercially available magnetite powder (Alchemie Research Centre, Mumbai, India) was ballmilled in a planetary ball mill (Pulverisette 5, FRITSCH, GMbH) in the presence of oleic acid. Using fractional sedimentation suspensions containing 3, 2, and 1 μm sized particles were obtained. The particles were almost spherical.[19]

Similarly suspensions of silica spheres (≈ 3 μm) were also prepared.

The requisite amount of the above suspensions was mixed with the diluted samples of ferrofluid and the mixture was homogenized by ultrasonification. No sedimentation was detected after the completion of observation. No change in the size of the scatterers was detected after the ultrasonification.

C. Experimental setup

Figure 1 shows the optical setup used in the investigation. A linearly polarized He-Ne laser (Jain Laser Tech., Mumbai, India) of wavelength $\lambda=633$ nm and 10 mW power was used as the light source. The beam was passed transversely through the magnetic field generated between the pole pieces of a small electromagnet energized by a constant current source. The field was found to be uniform between the pole gap. The electric vector of the incident beam was oriented parallel or perpendicular to the field direction by the Majorana effect.[27] It is known that the magnetically induced dichroism will be minimum when the E vector of the incident light is either parallel or perpendicular to the applied magnetic field. In the case of graphite dispersion the minima could be set up to an accuracy of 6′ arc by visually observing transmitted light through a second polarizer mounted on a graduated scale. The method is convenient to orient the polarization direction of the incident light for magneto-optical assembly.

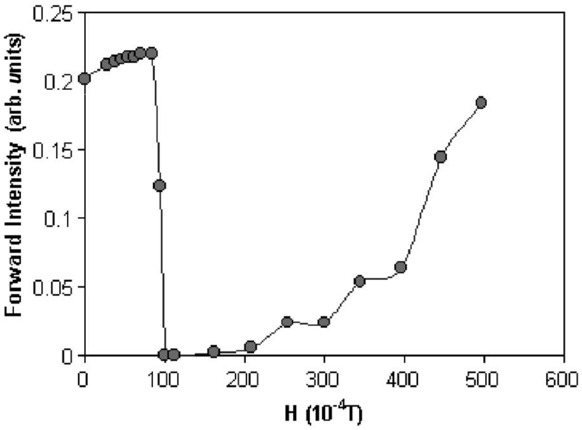

FIG. 2. Measurements of the forward scattered intensity as a function of applied magnetic field for the 3 μm sized magnetite spheres in ferrofluid.

The sample cell was made of a strain free glass having a 2 mm light path within the sample. The overall dimension of the cell was of height 2 cm and breadth 1 cm. This cell was filled with the sample under investigation and was mounted between the pole pieces of the magnet. Magnetically induced scattering patterns were recorded by a black and white CCD camera connected to a PC. All the investigations were carried out at room temperature (27 °C). Backward-scattering patterns were recorded by tilting the cell by $\xi \approx 10°$ to eliminate the directly reflected light from the cell surface.

For the photometric measurements a photodetector replaced the CCD. The output was amplified and fed to a current meter.

The detector was mounted on a traveling microscope placed at a distance of 40 cm from the sample cell and the

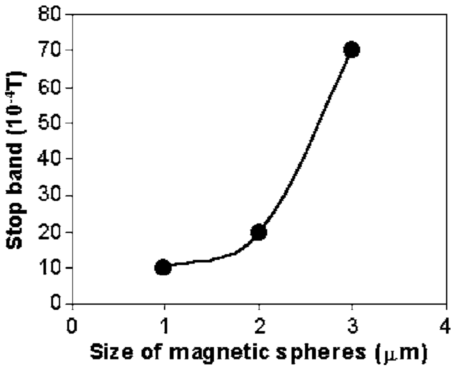

FIG. 3. Variation of stop band as a function of size of magnetic scatterers in ferrofluid.

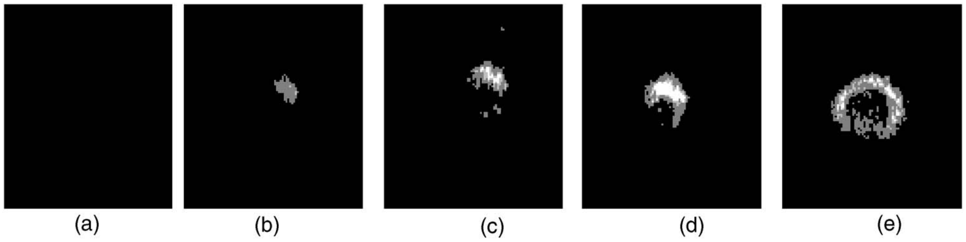

FIG. 4. CCD images of forward-scattering patterns after addition of different amount of silica spheres (~3 μm) in the mixture of ferrofluid and magnetite spheres. From (a)–(e) the addition of silica spheres are 0, 0.6, 1.0, 1.4, 2.4 mg, respectively.

intensity was measured as a function of horizontal distance of the microscope stage. This distance was converted into the cone angle.

III. RESULTS

A. Forward scattering

The ferrofluid was diluted 80 times and 6% of volume fraction of the 3, 2, and 1 μm size spheres were mixed, respectively, into the ferrofluid. All the measurements were carried out with these samples. The turbidity of these samples was found to be less than 0.1. Hence the effect of multiple scattering can be neglected.

In the previous paper[19] CCD images of magnetically induced forward scattering patterns were studied for four different sizes of magnetic scatterers. Photometric intensity of these patterns as a function of the field is plotted in Fig. 2. It is observed that the intensity drops to zero at a certain field H_{c1} and remain zero up to the field H_{c2}. Further increase in the field increases the intensity. The range of the field ($H_{c1}-H_{c2}$) within which the forward-scattered intensity remains zero may be called the *stop band of field* in analogy of *stop band of frequencies* in other photonic media.[6]. The band gap is found to decrease with a decrease in the size of the scatterers (Fig. 3). Like other photonic medium the magnetically induced photonic band gap (MPBG) is also affected by the addition of a small amount of nonmagnetic impurities. In the present case the impurity is 3 μm sized silica spheres. CCD images of the zero forward-scattering patterns after the addition of a different amount of impurities is shown in Fig. 4. Variation of the MPBG with concentration of the impurity is shown Fig. 5. The gap decreases with increase in the concentration and at certain concentration (2.4 mgm) it reduces to zero and no *zero forward-scattered* intensity was observed.

B. Backward scattering

Figure 6 shows the field induced backward scattering patterns for 3 μm size magnetic scatterers. For comparison the forward-scattered patterns observed at the same field are also shown. It is seen that at $H=100$ Oe both the forward as well as the backward scattered intensity disappear. The light appears to be localized in the medium. Similar patterns were also observed for the other sizes of the magnetic scatterers. Photometric measurements are shown in Fig. 7. It was observed that whenever the magnetically induced pattern disappears in the forward direction, in the backward direction it also disappears. Thus the propagation of light appears to stop at a certain critical field. However, in the backward direction no stop band was observed and a slight increase in the field increases the backscattered intensity by a large amount. At 200 Oe the back scattered intensity for 3 μm magnetite spheres is nearly 40 times larger than that of the forward scattered one and is nearly 1.6 times larger than the backscattered intensity at $H=0$ Oe. This is similar to the enhancement of coherent backscattering observed in other photonic materials but here it is induced by the applied magnetic field.[1]

Angular variation of this enhanced backscattering intensity is shown in Fig. 8. Here the angle 0° refers to the angle at which the intensity is maximum. Figure 9 shows the variation of the FWHM (full width at half maxima) ($\Delta\theta$) and the peak intensity with the size of the scatterers. It is observed that the width (height) decreases (increases) with the size of the scatterers.

IV. DISCUSSION

These results suggest that one can observe the localization of light by magnetically tuning the dispersion of the micron sized magnetic spheres in a ferrofluid. The medium offers a vivid demonstration of the complete disappearance of the light in the forward as well as in the backward direction. Variation of MPBG with the size and the backscattered cone parameters with the size are similar to that observed in other photonic media.

In the previous paper[19] occurrence of the magnetically induced *zero forward scattering* was explained on the basis

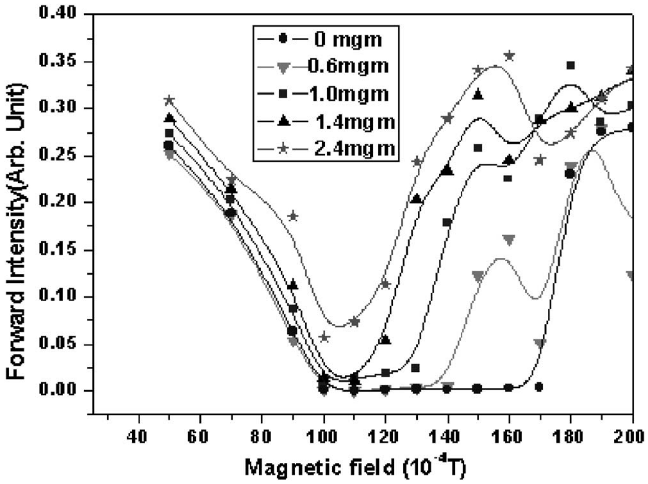

FIG. 5. Effect of addition of different amount of the impurity (i.e., silica spheres ~3 μm size) on the photonic MPBG. Line is drawn for clarity.

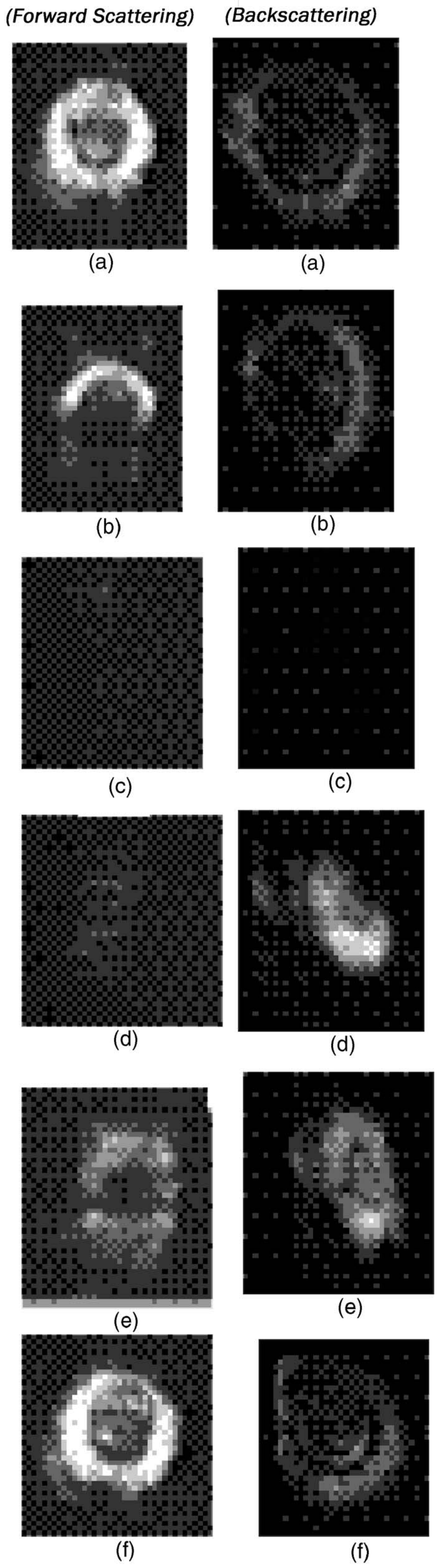

FIG. 6. Forward- and backward-scattering patterns for micron sized magnetite spheres in the ferrofluid (a) $H=0\ 10^{-4}$ T, (b) $H=50\times10^{-4}$ T, (c) $H=100\times10^{-4}$ T, (d) $H=200\times10^{-4}$ T, (e) $H=250\times10^{-4}$ T, (f) $H=500\times10^{-4}$ T. Complete vanishing of forward / backward-scattering light is evident in (c).

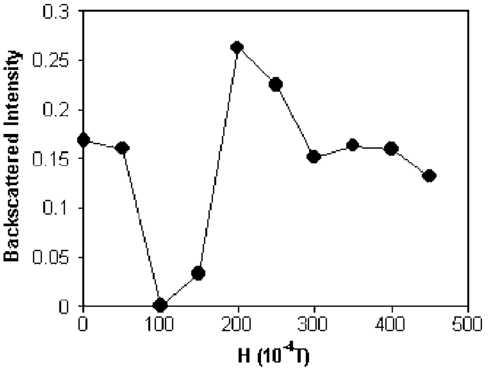

FIG. 7. Variation of the backscattered intensity with the field.

of a special case treated by Kerker *et al.* in their theory of electromagnetic scattering by a magnetic sphere.[17] Assuming single and independent scattering and considering the fact that ε_f and μ_f of the ferrofluid will vary with the applied field disappearance of the forward-scattering patterns were explained by assuming the Rayleigh-Gans approximation to be valid for the micron sized magnetic spheres. But this model cannot explain the results described in the above section. According to the Kerker's theory in the small particle limit the scattered intensity in a direction θ for the two orthogonal states of polarization are given by (Ref. 17)

$$I_1(\theta)=\frac{\lambda^2}{4\pi^2r^2}\left(\frac{2\pi a}{\lambda}\right)^6\left[\left(\frac{\varepsilon-1}{\varepsilon+2}\right)+\left(\frac{\mu-1}{\mu+2}\right)\cos\theta\right]^2\sin^2\phi, \tag{1}$$

$$I_2(\theta)=\frac{\lambda^2}{4\pi^2r^2}\left(\frac{2\pi a}{\lambda}\right)^6\left[\left(\frac{\varepsilon-1}{\varepsilon+2}\right)\cos\theta+\left(\frac{\mu-1}{\mu+2}\right)\right]^2\cos^2\phi. \tag{2}$$

When $\varepsilon=\frac{4-\mu}{2\mu+1}$ and $\theta=0$, $I_1(0°)$ and $I_2(0°)=0$. But when $\theta=180°$ and $\varepsilon=\frac{4-\mu}{2\mu+1}$,

$$I_1(180°)=\frac{\lambda^2}{4\pi^2r^2}\left(\frac{2\pi a}{\lambda}\right)^6\left[\left(\frac{\varepsilon-1}{\varepsilon+2}\right)+\left(\frac{\mu-1}{\mu+2}\right)\right]^2\sin^2\phi \neq 0. \tag{3}$$

Similarly, $I_2(180°)\neq0$.

In the present observation both $I_1(0°)$ and $I_1(180°)$ reduces to zero at the field $H=H_c$.

It is interesting to note that when $\varepsilon=\mu=-2$ Kerker's conditions for the zero forward as well as the zero backward scattering are satisfied.[17] However, the relative magnetic permeability with respect to the ferrofluid is unlikely to be negative. Hence one cannot explain the occurrence of the vanishing of light by means of Kerker's predictions. Further the observed MPBG also cannot be explained on the basis of the theory given by Kerker *et al.*

The magnetic field induced changes in the diffraction pattern produced by a focused laser beam on a horizontal layer of magnetic fluid was earlier studied by Luo *et al.*[28] They have shown that when the field is applied parallel to the laser beam a convective instability sets in resulting in the changes

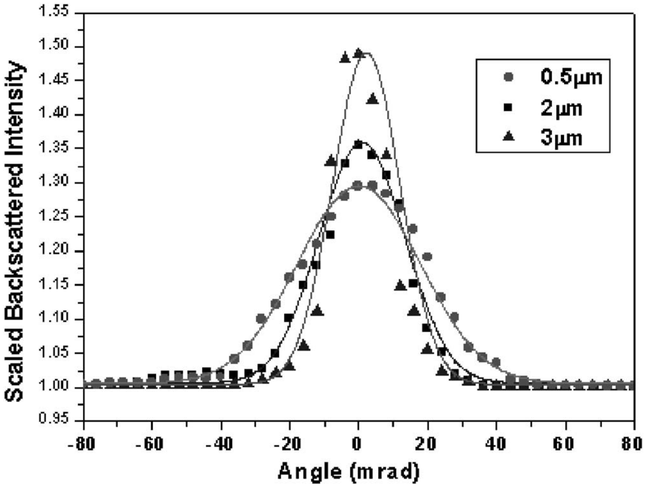

FIG. 8. Angular variation of the enhanced backscattered intensity for the micron sized magnetite spheres in ferrofluid. The absolute value of the angle is 10° corresponds to the relative value 0. Line shows the Gaussian fit to the experimental data.

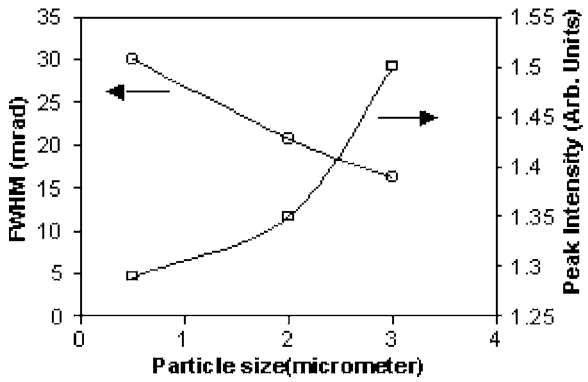

FIG. 9. Variation of the FWHM (full width at half maxima) and the peak intensity in backscattered direction with the size of the magnetite spheres in ferrofluid.

in the shape of the diffraction patterns. In the present case since a parallel beam of light is used the Soret effect, if any, will be negligible. The forward and backward patterns are generated due to the Mie scattering. Further the field direction in the present case is transverse to the light beam. Hence convective instability is not likely to be responsible for the observed effects.

Wiersma *et al.* have investigated the optical transport properties of complex photonic structures ranging from ordered photonic crystals to disordered strongly scattering materials.[29] Polydispersity is assumed to control the amount of order/disorder. In the present system the same may be assumed to be controlled by the magnetic field. Under this situation it is likely that the system exhibits anisotropic structures. It may be remarked here that formation of such structures in a dispersion of magnetic microspheres in a ferrofluid are shown by Skjeltorp *et al.*[30] The cumulative effect due to Mie scattering by magnetic spheres will depend upon how the spheres orient with respect to each other. Such a partially oriented dispersed system may exhibit a stop band of field. Further experimental as well as theoretical work on Mie scattering by magnetic spheres dispersed in a ferrofluid is needed to reach a definite conclusion.

V. SUMMARY

The present investigations demonstrated a vanishing of light in the forward as well as in the backward direction when a critical magnetic field is applied to a dispersion of magnetic scatterers in a ferrofluid. Upon increasing the field from this critical value enhancement of the backscattered intensity is observed. In the forward direction a magnetically tunable phonic band gap is observed which is affected by the presence of impurities. The backward scattered cone angle and cone height was found to depend on the size of the scatterers. These effects are similar to that observed in other photonic media. The technique of magnetic tuning is simple and requires only a moderate magnetic field and hence will be useful to develop photonic devices.

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