Experimental Evidence of Zero Forward Scattering by Magnetic Spheres

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(Received 8 December 2005; published 28 March 2006)

Magnetically induced diffraction patterns by micron sized magnetic spheres dispersed in a ferrofluid disappear at a certain critical magnetic field. This critical field is found to depend on the concentration of the ferrofluid and on the volume of the magnetic spheres. We attribute this effect to the zero forward scattering by magnetic spheres as predicted by Kerker, Wang, and Giles [J. Opt. Soc. Am. **73**, 765 (1983)]. We suggest that such a dispersion can be used to study the optical analogues of localization of electrons in condensed matter, the Hall effect, and the anisotropic diffusion, etc. The combination of the micron sized magnetic spheres and the ferrofluid will also be useful to design magnetically tunable photonic devices.

DOI: 10.1103/PhysRevLett.96.127402 PACS numbers: 78.20.Ls, 42.70.Qs, 147.65.Cb, 75.50.Mm

Analogies between the propagation of electromagnetic waves in strongly scattering media and that of electrons in condensed matter have attracted a good deal of attention [1–5]. The photonic Hall effect, photonic magnetoresistance, localization of light, and anisotropic diffused coefficients are the optical analogue of the corresponding effects in condensed matter. Recently, switchable Bragg diffraction, for liquid crystals in colloid templated materials, has been observed by Mach *et al.* [6]. They have also studied anisotropic weak localization of light in liquid crystals [7]. Pinheiro *et al.* [8] have shown that, when scatterers are ferromagnetic, it may favor the observation of localization of electromagnetic waves. It has been suggested that if soft ferrites are used as the scatterers it may be easy to observe the localization of the incident microwave radiation near the Curie-Weiss temperature [9]. Effects of magnetic field on light scattering is also studied by van Tiggelen *et al.* [10]. Single scattering of electromagnetic waves by magnetic spheres was studied earlier by Kerker *et al.* [11]. Theoretically several unusual scattering effects were predicted. In the small particle limit, forward scattering will become zero when the scatterers satisfy the condition between the relative electric permittivity (ε) and the relative magnetic permeability (μ). Such effects are rather difficult to observe at optical frequencies under normal conditions since in this range $\mu \approx 1$ [11]. Zero forward scattering is also possible when propagation of electromagnetic radiation takes place through a metal film in which holes are filled with magnetic colloidal particles. Helseth has suggested that in such medium plasma frequency can be tuned by an external field [12]. He has predicted a shift in the transmission peaks of microwaves on increasing the field. It may be possible to extend the analysis in case of the visible or infrared region. Experimental verification of the above unusual effect has not been so far reported. Here we propose that if the magnetic scatterers are dispersed in a ferrofluid, it is possible to vary the relative permittivity as well as the permeability of the scatterers by varying an externally applied magnetic field, and it is possible to tune the condition for zero forward scattering. We report the first experimental evidence of the zero forward scattering by magnetite spheres dispersed in a ferrofluid. Such dispersion can also serve as a medium to study other photonic effects.

When a beam of electromagnetic radiation passes through a medium, part of the energy of radiation is removed and the remaining is transmitted through the medium. The fraction of the removed part may be absorbed in the medium and the remaining fraction is scattered in the same direction, as well as in all other directions. The amount of absorbed and scattered energy depends upon the nature of the scatterers, as well as the wavelength of the incident radiation in the medium (λ). When the scatterers are magnetic spheres dispersed in a homogeneous medium, the intensity and state of polarization of the scattered beam depend on size, the *relative electric permittivity* ($\varepsilon = \varepsilon_s/\varepsilon_m$), and the *relative magnetic permeability* ($\mu = \mu_s/\mu_m$) [11], where subscripts "s" and "m" stands for scatterers and medium, respectively. Usually the medium is considered to be free space or an isotropic dielectric nonabsorbing medium with $\mu_m = \mu_0$, which leads to the real refractive index $m_m = \sqrt{\varepsilon_m \mu_0}$. The relative refractive index of the scatterers will be $m = m_s/m_m = \sqrt{\mu\varepsilon}$, where $\mu = \mu' + i\mu''$ and $\varepsilon = \varepsilon' + i\varepsilon''$ are the complex relative magnetic permeability and electric permittivity, respectively. Then using Mie formalism, the scattered intensities of two polarized components are given by [8,9].

$$I_1 = (\lambda^2/4\pi^2 r^2)|S_1|^2 \sin^2\phi, \tag{1}$$

$$I_2 = (\lambda^2/4\pi^2 r^2)|S_2|^2 \cos^2\phi. \tag{2}$$

Where r is the distance between the scatterers and the observer, ϕ is the angle between the electric vector of the incident wave and the scattering plane, and I_1 and I_2 are the intensities for the two orthogonal states of polarization, viz., for the electric vector perpendicular and parallel to the scattering plane.

Expressions of the scattering amplitudes $S_{1,2}$ in terms of the Mie coefficients a_n and b_n and Legendre polynomials π_n $(\cos\theta)$ and τ_n $(\cos\theta)$ are given by Kerker *et al.* [11]. For small particles, only first term of the coefficients are significant. Using these it is shown that when

$$\varepsilon = \frac{4 - \mu}{2\mu + 1} \tag{3}$$

and $\theta = 0$ both $I_1(0^0)$ and $I_2(0^0) = 0$. Hence, the intensity of forward scattering becomes zero.

This condition is difficult to satisfy (at least at optical frequencies) for the magnetic scatterers surrounded by isotropic dielectric nonmagnetic medium. The condition to observe zero forward scattering gets modified when the scatterers are dispersed in a ferrofluid. Under the influence of the magnetic field, ferrofluid exhibits induced anisotropy and the medium becomes birefringent [13,14]. Consequently, the condition for zero forward scattering [Eq. (3)] will be different for parallel and perpendicular states of polarization.

Recently we have studied field induced diffraction patterns and the induced extinction in a magneto-rheological suspension of magnetite in a ferrofluid [15]. It was observed that the pattern, as well as the extinction, changes significantly with an increase in the field. The pattern disappears at a critical field. An attempt was made to interpret the results on the basis of dipole scattering and geometrical shadow approximation [16]. But this was unsuccessful. Similarly competition between torques due to the permanent magnetic moment and magnetic hole also failed to explain the observed effect. The present investigation clearly reveals that the disappearance of the Fraunhoffer diffraction occurs only when magnetic spheres are dispersed in a ferrofluid. We attribute this effect to the zero forward scattering by magnetic spheres.

In this Letter we describe these investigations. It is to be noted here that the micron sized magnetic particles are not smaller than the wavelength of light. So the Rayleigh theory is not strictly applicable. But if we consider the fact that scattering coefficients (the sum of conservative and consumptive scattering) of Rayleigh scatterers and Rayleigh-Gans scatterers are nearly equal in the case of absorbing particles, then as a first approximation, our analysis can remain valid.

A linearly polarized He-Ne laser beam of 10 mW power was used as the light source. This beam was passed through the sample contained in a 0.2 cm optical path length glass cell. The cell was placed between the pole pieces of a small electromagnet energized by a constant current power supply. The polarization vector of the incident light was oriented parallel or perpendicular to the transverse electromagnetic field by the magneto-optical technique [17]. Forward scattering patterns as a function of the applied magnetic field were recorded by CCD camera connected to a personal computer.

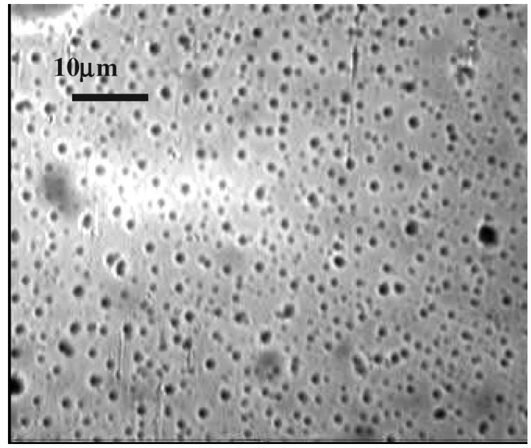

FIG. 1. A typical microscope picture of 1.0 μm.

The micron sized magnetite particles used in the study were found to be almost spherical. A typical microscope picture of 1.0 μm diameter particles is shown in Fig. 1. These particles were dispersed in the kerosene based magnetic fluid and concentration was kept low enough to avoid multiple scattering [15].

Figs. 2 and 3 show the diffraction patterns for 3 μm size magnetite particles dispersed in the ferrofluid for the electric vector $\mathbf{E}$ of the incident light parallel and perpendicular to the applied magnetic field $\mathbf{H}$, respectively. The diffraction pattern exhibits isotropic scattering due to the micron sized magnetic particles for $H = 0$. On increasing the magnetic field, the diffraction patterns for both the polarization states, i.e., $\mathbf{E} \parallel \mathbf{H}$ and $\mathbf{E} \perp \mathbf{H}$ get modulated. When $\mathbf{E} \perp \mathbf{H}$ zero forward scattering was observed for a critical field of $\sim$100 G [Fig. 3(d)]. While no such zero forward scattering was observed for $\mathbf{E} \parallel \mathbf{H}$ (Fig. 2). As

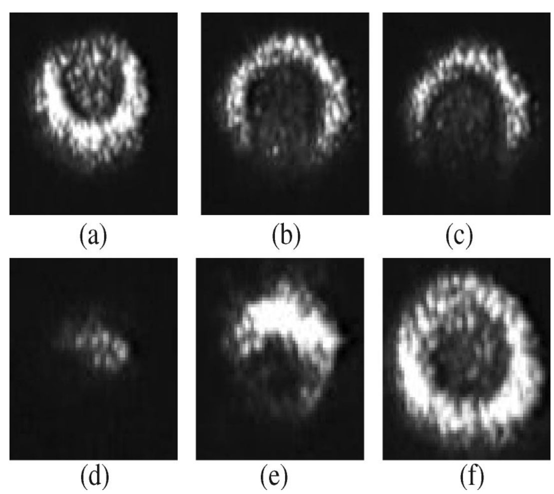

FIG. 2. Diffraction pattern for the electric vector parallel to the applied magnetic field in the case of 3 μm magnetic spheres dispersed in a ferrofluid (28% concentration); (a) $H = 0$ G, (b) $H = 30$ G, (c) $H = 60$ G, (d) $H = 100$ G, (e) $H = 200$ G, (f) $H = 500$ G.

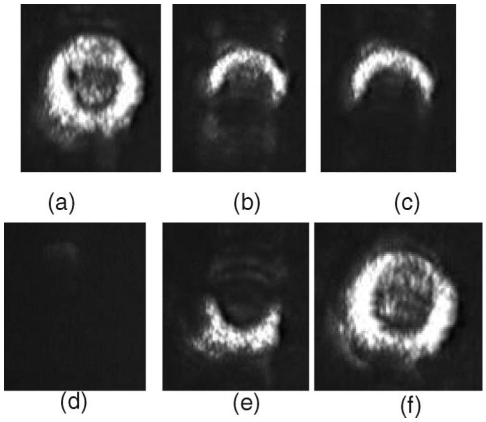

FIG. 3. Diffraction pattern for the electric vector perpendicular to the applied magnetic field in the case of 3 μm magnetic spheres dispersed in a ferrofluid (28% concentration); (b) $H = 30$ G, (c) $H = 60$ G, (d) $H = 100$ G, (e) $H = 200$ G, (f) $H = 500$ G. Zero forward scattering is observed for $H = 100$ G (d).

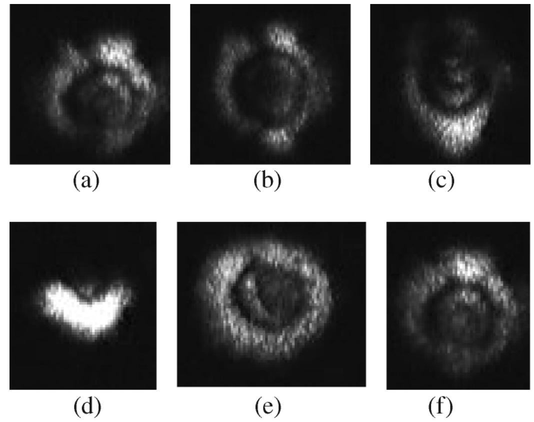

FIG. 5. Diffraction pattern for the electric vector perpendicular to the applied magnetic field in the case of 3 μm magnetic spheres dispersed in kerosene (a) $H = 0$ G, (b) $H = 30$ G, (c) $H = 60$ G, (d) $H = 100$ G, (e) $H = 200$ G, (f) $H = 500$ G.

discussed earlier, a ferrofluid becomes birefringent under the influence of the applied magnetic field. Hence $\varepsilon_{\parallel}$ and $\varepsilon_{\perp}$ will be different for the two orthogonal polarization states. The condition for zero forward scattering is found to be satisfied only for $\mathbf{E} \perp \mathbf{H}$, i.e., I_1 and not for $\mathbf{E} \parallel \mathbf{H}$, i.e., I_2. This also confirms the deduction by Kerker *et al.* [11].

In order to verify that the zero forward scattering occurs only when the magnetic scatterers are dispersed in the ferrofluid we have recorded diffraction patterns for the following samples. (i) The medium is ferrofluid but the dispersed phase is nonmagnetic micron sized silica particles. (ii) Dispersed particles are micron sized magnetite particles and the medium is a magnetically passive liquid, i.e., kerosene.

The zero forward scattering was not observed in either of the samples for $\mathbf{E} \perp \mathbf{H}$ as well as $\mathbf{E} \parallel \mathbf{H}$. Typical diffraction patterns are shown for $\mathbf{E} \perp \mathbf{H}$ (Figs. 4 and 5). This confirms that the zero forward scattering occurs only when the dispersed phase is magnetic and the medium is ferrofluid.

In the low concentration regime (dipole-dipole interaction is negligible) permittivity and permeability will vary linearly with the concentration. Hence it is expected that the critical field, H_c, at which the zero forward scattering is observed, will also vary linearly with concentration. We have carried out the study for three different concentrations of the original fluid. Figure 6 shows the plot of H_c vs concentration. The linearity confirms the above deduction. Similarly, H_c also depends on the volume of the micron sized magnetic scatterers (Fig. 6). The dependency of H_c on concentration and on the size of the scatterers is being analyzed on the basis of the effective medium theory given by Maxwell-Garnett [18]. The present investigations demonstrate that it is possible to observe magnetically induced zero forward scattering at optical frequency with a proper

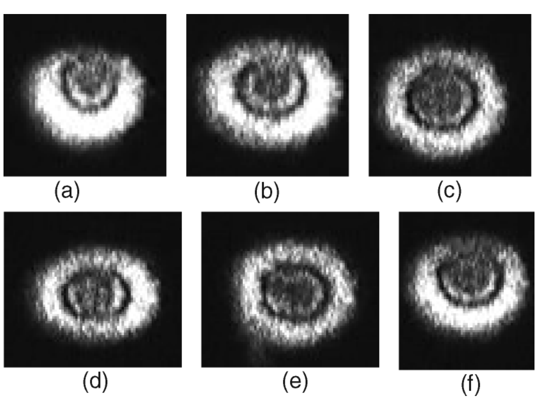

FIG. 4. Diffraction pattern for the electric vector perpendicular to the applied magnetic field in the case of 2–3 μm silica spheres dispersed in a ferrofluid (28% concentration) (a) $H = 0$ G, (b) $H = 30$ G, (c) $H = 60$ G, (d) $H = 100$ G, (e) $H = 200$ G, (f) $H = 500$ G.

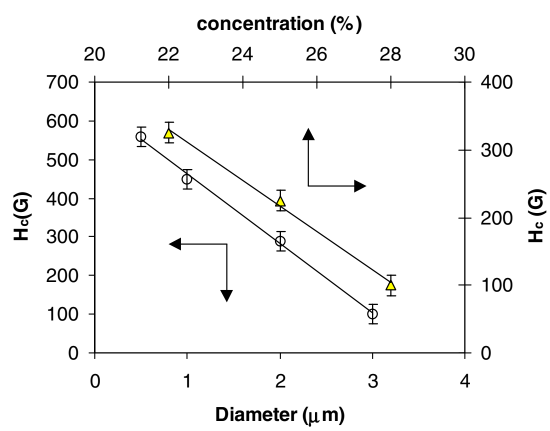

FIG. 6 (color online). Dependency of critical magnetic field (H_c) for zero forward scattering on concentration and on the size of the scatterers dispersed in a ferrofluid.

combination of ferrofluid and magnetic scatterers. This combination will be useful to study the multiple scattering effects like photonic Hall effect, localization, and anisotropic diffusion of light. It may also help to design magnetically tunable devices like photonic systems.

The work was carried out under the emeritus Scientist Scheme of CSIR, India [Scheme No. 21 (0537)/02/EMR-II]. R. V. U. is thankful to GUJCOST. R. V. U., R. P., and R. D. are thankful to DST for providing support in terms of DST-M-15 Project.

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