Nonreciprocal light diffraction by a lattice of magnetic vortices

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We report an experimental study of the optical properties of a two-dimensional square lattice of triangle Co and CoFe nanoparticles with a vortex magnetization distribution. We demonstrate that the intensity of light scattered in the diffraction maxima depends on the vorticity of the particles' magnetization and can be manipulated by applying an external magnetic field. The experimental results can be understood in terms of simple phenomenological consideration.

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Considerable achievements in microtechnologies and nanotechnologies open new possibilities in fabrication of artificial nanomaterials (metamaterials) with novel interesting optical properties. Planar metallic structures attract attention owing to many interesting effects appearing in them, specifically, the enhanced magneto-optical effects,[1–3] extraordinary light transmission through the subwavelength holes,[4] and effective generation of the second harmonic.[5–7] Special attention is paid to planar structures consisting of chiral elements.[8–13] Such structures are ordinarily a two-dimensional regular lattice of nonmagnetic particles that do not possess a reflection symmetry (in the plane perpendicular to the sample surface) and can be characterized by a *pseudovector (axial vector)*. This circumstance determines a phenomenon of asymmetric polarization conversion.[14] In this paper, we explore a magnetic planar chiral structure. As will be shown in the following, such structures can be characterized by a *polar vector* that changes its sign under the time reversal. This causes nonreciprocal optical effects[15–17] that can not exist in nonmagnetic structures.

In our work, we investigate regular lattices of particles with the vortex magnetization distribution. In contrast to a nonmagnetic planar chiral structure, the spatial inversion symmetry is broken in a vortex particle due to nontrivial magnetization distribution. The vorticity direction can be manipulated with the tip of magnetic-force microscope,[18] by uniform external magnetic field,[19] or by applying an electric current.[20] Shaping the magnetic particles as triangles, one can get all the particles to have the same vorticity and thus the same planar chirality by means of a uniform magnetic field. In this paper, we report an experimental observation of nonreciprocal effects in light diffraction by a two-dimensional lattice of magnetic vortices.

We begin with some phenomenological arguments in favor of nonreciprocal light diffraction by a vortex particle. If one considers the light-scattering cross section summed over the polarization of incident and diffracted light, the reciprocity law takes a simple form

$$\sigma[\mathbf{k},\mathbf{k}',\mathbf{M}(\mathbf{r})] = \sigma[-\mathbf{k}',-\mathbf{k},-\mathbf{M}(\mathbf{r})], \quad (1)$$

where σ is the differential cross section for the diffracted light, $\mathbf{k}$ and $\mathbf{k}'$ are the wave vectors of the incident and diffracted beams, and $\mathbf{M}(\mathbf{r})$ is the magnetization spatial distribution. The term "nonreciprocal effect" implies one of the two equivalent inequalities

$$\sigma[\mathbf{k},\mathbf{k}',\mathbf{M}(\mathbf{r})] \neq \sigma[-\mathbf{k}',-\mathbf{k},\mathbf{M}(\mathbf{r})], \quad (2)$$

$$\sigma[\mathbf{k},\mathbf{k}',\mathbf{M}(\mathbf{r})] \neq \sigma[\mathbf{k}',\mathbf{k},-\mathbf{M}(\mathbf{r})]. \quad (3)$$

For systems without the center of inversion, the scattering cross section may contain the term $[(\mathbf{k}+\mathbf{k}')\cdot\mathbf{C}]$, where $\mathbf{C}$ is a vector. It is linear in the wave-vector term, which leads to nonreciprocal effects described by Eqs. (2) and (3). According to the reciprocity law, for systems without spatial inversion, the $\mathbf{C}$ vector should be a *polar vector* that also changes its sign under the time reversal. For a magnetic scatterer of centrosymmetrical shape made of a centrosymmetrical material, $\mathbf{C}$ can be chosen in the simplest form $\mathbf{C} = \alpha\langle[\mathbf{r}\times\mathbf{M}(\mathbf{r})]\rangle$, which is a toroidal moment of the particle associated with the magnetic vorticity[21] (the square brackets mean the spatial averaging over the scatterer, α is a constant). It follows from the above that the scattering of unpolarized light by a particle with the vortex magnetization distribution is nonreciprocal, its contribution being dependent on the vorticity:

$$\sigma[\mathbf{k},\mathbf{k}',\mathbf{M}(\mathbf{r})] = \cdots + \alpha[(\mathbf{k}+\mathbf{k}')\cdot\langle[\mathbf{r}\times\mathbf{M}(\mathbf{r})]\rangle]. \quad (4)$$

We carried out experimental investigations of light diffraction by the lattices of magnetic vortices in order to confirm the fact of such a contribution in the scattering cross section.

Two-dimensional (2D) arrays of 30-nm-thick polycrystalline triangular Co and CoFe dots were fabricated by the electron beam lithography and lift-off technique on the surface of an amorphous SiO_2 plate. The details of the technological procedures can be found in Ref. 22. The dots are arranged in a 400×400 μm^2 area and disposed in a square lattice with a period of 1.4 μm. The period makes it possible to observe the diffraction maxima of the HeNe laser beam ($\lambda = 632$ nm) used in our measurements. The size of the particles along a triangle side is 0.7 μm [Fig. 1(a)]. This size was set based on two competing requirements: (1) to obtain the maximum possible volume of the magnetic material and (2) to avoid a significant magnetostatic interaction of particles because it may affect their magnetization state.[22]

In a zero external field, the ground magnetic state of a triangle particle of dimensions as mentioned above is the vortex state [Figs. 1(b) and 1(c)]. The left- and right-hand vortices have different signs of vorticity and, thus, should have different light-scattering cross sections, in accordance

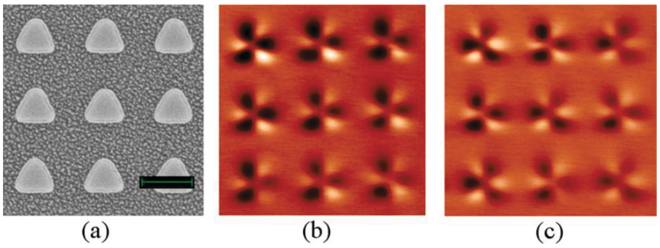

FIG. 1. (Color online) (a) Scanning electron microscope (SEM) image of the lattice of the cobalt triangles, the scale bar length is 1 μm. (b) MFM image of the remanent states after magnetizing along the base of the triangles. All magnetic vortices demonstrate the same direction of the vorticity. (c) MFM image of the remanent states after magnetizing along the height of the triangles. The vortices with both CW and CCW vortices are presented.

with Eq. (4). The magnetization hysteresis loop of the lattice of Co triangles in the case when external field is applied along the triangle base has a shape typical of the particles with magnetic vortices [Fig. 2(a)].[23] These data were obtained by the magneto-optical Kerr effect (MOKE) measurement in the meridional configuration at room temperature. In a high positive magnetic field, all particles are magnetized uniformly. When the magnetic field is decreased from saturation, a magnetic vortex nucleates, which is accompanied by an abrupt decrease in magnetization. If the field is directed along the base of the triangle to the right [Fig. 2(c)], only the counterclockwise (CCW) vortices enter the particles. It is a direct consequence of the particles having a noncentrosymmetric triangular shape. The magnetization states were verified in the magnetic force microscopy (MFM) investigations. Depending on a sample 90%–100% of particles were found to be in the same vortex state in a zero field after such a procedure [Fig. 1(b)]. In a high negative field, all particles are uniformly magnetized in the field direction again. When the magnetic field increases from high negative values, the magnetization vorticity in the particles turns clockwise (CW). So, by applying a uniform external field, we can synchronically manipulate the vorticity of all particles.[19] If the magnetic field is oriented along the triangle height, the shape of the hysteresis loop is practically the same. Yet, the probability of the CW and CCW vortex nucleation in this case is equal and there is a mixture of vortices with both vorticities in a zero external field [Fig. 2(c)].

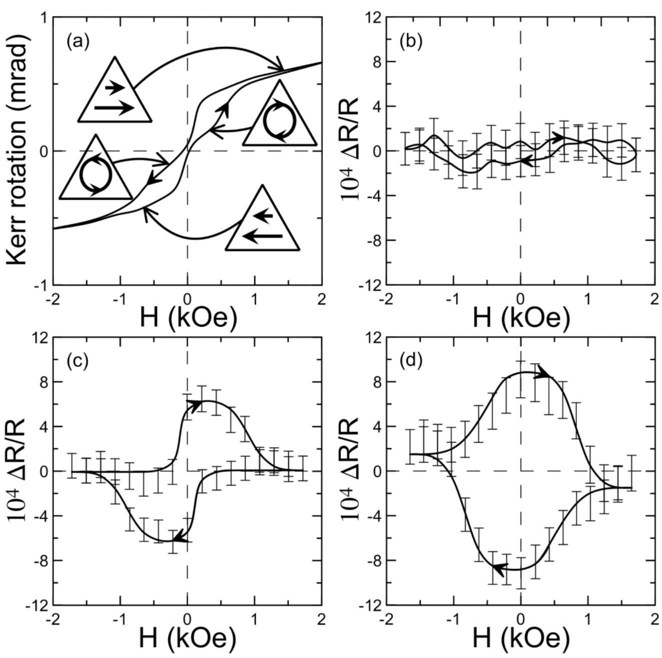

FIG. 2. (a) In-plane magnetization curve of the lattice of ferromagnetic triangles measured by the magneto-optic Kerr rotation. Magnetic field is applied along the triangles' side. (b)–(d) Relative change of the light intensity [$\Delta R = R(H) - \langle R(H)\rangle$, square brackets mean averaging over the magnetization cycle, $R(H)$ is the intensity of the diffracted light, H is the magnitude of the external magnetic field] diffracted in $(-1,0)_{\rm tr}$ maximum as a function of the applied magnetic field. The incident angle is 5°. (b) The field is directed along the triangles' height. (c) The field is directed along the triangles' side. Incident wave is s polarized. (d) The field is directed along the triangles' side. Incident wave is p polarized. Error bars represent the experimental data; solid lines are guides for eyes showing the direction of the hysteresis loop traversal during the magnetizing cycle. The magnetization distributions corresponding to the hysteresis loop segments are represented schematically (view in the z direction).

The geometry of the optical measurements is depicted in Fig. 3. The sample lies in the (x,y) plane with the lattice vectors oriented along the x and y axes. The averaged toroidal moment $\langle[\mathbf{r} \times \mathbf{M}(\mathbf{r})]\rangle$ is parallel to the z axis in this geometry. The sample was irradiated with a laser beam propagating in the (z,x) plane at the angles 5°–40° with respect to the z axis. The intensity of the diffracted light was measured in four diffraction maxima lying in the same plane (dashed lines in Fig. 3). These are $(\pm 1,0)$ maxima both for the transmitted and reflected light. The external magnetic field was directed in the plane of the sample parallel to the x axis. During the measurement, the data were taken as a function of the magnetic field in order to generate a hysteresis loop. The measurements were carried out separately for the s and p polarizations of the incident light. To compare the experimental results with the prediction Eq. (4), the diffracted intensity for different polarizations should be summed over.

To check the phenomenological predictions [Eq. (4)], we investigate the dependence of the intensity of light scattered in the diffraction maxima on the direction of a magnetic particle vorticity. Measurements for different incident angles to explore the diffraction maxima with the positive and negative values

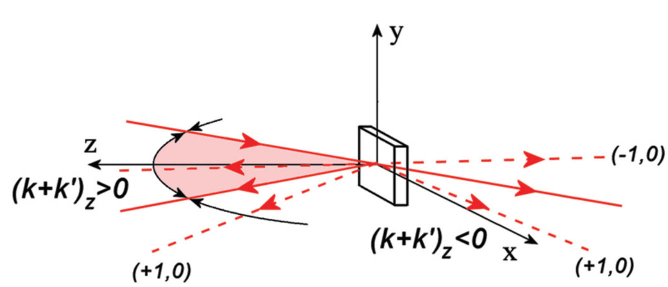

FIG. 3. (Color online) The geometry of the experiment. Solid lines are incident, transmitted, and specularly reflected beams, dashed lines represent diffracted beams. The gray-colored (red online) segment of the XZ plane corresponds to the directions with $(k+k')_z > 0$.

of $k_z + k'_z$ were also carried out. The main results of the experiment are summarized in the following:

(i) During the magnetizing cycle (starting from the saturation field along the x axis), the magnetic particles sequentially pass the following states: homogeneous (single domain) → CCW vortex → homogeneous → CW vortex. The difference in the intensity magnitude of light diffracted by the CW and CCW vortices leads to appearance of a hysteresis loop [Figs. 2(c) and 2(d)]. Indeed, in a zero field we have different diffracted intensities for the CW and CCW vortices. The shape of the hysteresis loop is different for the s and p polarizations, but the direction of the hysteresis curve traversal (i.e., the sign of the effect) does not depend on polarization. Thus, if we sum the diffracted intensities over the incident polarizations, the hysteresis loop will not disappear and we will have the polarization independent part of the effect [described by Eq. (4)].

(ii) If the magnetizing field is directed along the height of the triangles, no change in the intensity of the light scattered in the diffraction peaks is observed. Indeed, the CW and CCW vortices have equal probability of appearance in this situation and, consequently, their number in the system is the same. Hence, nonreciprocal effects disappear.

(iii) For the incident angle of 5°, the effect has the same sign for the (±1,0) maxima both in reflection and transmission. This can be explained by the fact that in this geometry the value of $k_z + k'_z$ is always negative (see Fig. 3). If the incident angle is 30°, the sum $k_z + k'_z$ becomes positive for the $(-1,0)_{\text{refl}}$ diffraction maximum, but remains negative for the $(+1,0)_{\text{refl}}$ diffraction maximum (as in the previously described experiments with the incident angle of 5°). Simultaneous measurements of the intensity of light scattered in these diffraction maxima with the external magnetic field applied along the base of the triangle were carried out. We found out that the intensity of the diffracted light in these two maxima changes with a different sign for the same direction of the vorticity. It is manifested in opposite directions of the hysteresis loop circumvention in these cases (Fig. 4).

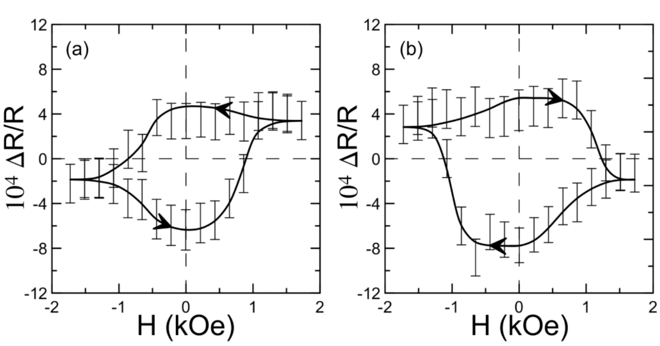

FIG. 4. Relative change of the intensity of the light diffracted (a) in $(-1,0)_{\text{ref}}$ maximum ($k_z + k'_z > 0$) and (b) in $(+1,0)_{\text{ref}}$ maximum ($k_z + k'_z < 0$) measured simultaneously in a single run as a function of an applied magnetic field. The incident angle of the s-polarized light is 30°. Error bars represent experimental data, and solid lines are guides for eyes showing the direction of the hysteresis loop traversal. The change of the hysteresis from counterclockwise-to-clockwise type with the change of the sign of $(k_z + k'_z)$ is evident.

The experiments demonstrate that the intensity of the light diffracted by the vortex lattice depends on the scalar product $[(\mathbf{k} + \mathbf{k}') \cdot \langle[\mathbf{r} \times \mathbf{M}(\mathbf{r})]\rangle]$, i.e., it has nonreciprocal character. Thus, in spite of the zero average magnetization of particles in a zero external field, the existence of the magnetic vortex is manifested through the intensity of the diffracted light.

We propose two possible mechanisms causing appearance of the nonreciprocal term in the scattering cross section. The first one is the excitation of an electric dipole in a vortex particle under the influence of uniform magnetic field of the incident wave. It is well known that a magnetic field induces a vortex eddy current in a conductive particle. The influence of the magnetic vortex on this current leads to an additional contribution in the electrodipole moment of the particle. The sign of this contribution depends on the vorticity. The other possible mechanism is the excitation of an electric quadrupole moment in a particle with the vortex magnetization distribution under the impact of the uniform electric field of the incident wave. The quadrupole electric moment appears due to the anomalous Hall effect and nonuniform magnetization distribution. Although the quadrupole effect is usually small compared to the dipole one, it can not be neglected here. Indeed, the contribution of the above-mentioned addition to the dipole moment described above is of the same order of magnitude with respect to the ratio of a particle size to wavelength due to the fact that it is caused by the spatial dispersion (the magnetic field of the wave). The phase of the quadrupole moment oscillation depends on the vorticity of a particle. The interference of the waves radiated by the quadrupole and dipole leads to a vorticity-dependent contribution in the scattering cross section.

As can be seen from Figs. 4 and 2, the intensity of the diffracted light is different for the saturation fields of opposite direction. This means that the term linear in magnetization $\alpha_{i,j}(k + k')_i M_j$ appears in the scattering cross section. Here, $\alpha_{i,j}$ is a pseudotensor. The lattice of the triangles has only the reflection mirror plane (y,z) (here x is directed along the triangles' base, y is along the triangles' height, both x and y are along the lattice vectors, z is perpendicular to the sample surface). So, the pseudotensor components $\alpha_{x,y}, \alpha_{x,z}, \alpha_{y,x}, \alpha_{z,x}$ are nonzero. This is in agreement with the experimental data. From Figs. 2(b) and 2(d), one can see that the effect is zero when the magnetic field is directed along the y axis ($\alpha_{z,y} = 0$) and it is nonzero when the field is along the x axis ($\alpha_{z,x} \neq 0$). Also, the effect changes sign when the z projection of vector $\mathbf{k} + \mathbf{k}'$ changes sign (see Fig. 4).

In conclusion, light diffraction by the two-dimensional square lattice of triangle Co and CoFe nanoparticles in the vortex magnetic state has been investigated. The peculiarity of the system is that all particles have the same vorticity that can be manipulated by applying a uniform external magnetic field. We observed a nonreciprocal intensity effect that consists in the dependence of the diffracted intensity on the particles' vorticity. The observed effect could be described by the phenomenological expression $\alpha[(\mathbf{k} + \mathbf{k}') \cdot \langle[\mathbf{r} \times \mathbf{M}(\mathbf{r})]\rangle]$. Possible microscopic reasons underlying the effect have been also discussed.

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