Chiral Symmetry Breaking in Magnetic Thin Films and Multilayers

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A phenomenological theory of chiral symmetry breaking in magnetic nanostructures is developed considering induced, inhomogeneous chiral interactions (Dzyaloshinsky-Moriya–type). Application of the theory to films and multilayers with in-plane and out-of-plane magnetization predicts modulated and two-dimensional localized patterns (vortices). These new classes of magnetic patterns are intrinsically stable and localized on nanometer scale. Various experimental observations agree qualitatively with structures derived from this theory.

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Chiral asymmetry is ubiquitous in nature from cosmic objects (spiral galaxies and polarized stellar light) to the enigmatic homochirality of biomolecules in all forms of terrestrial life, and the parity violation in particle physics [1]. Chirality is of central importance in modern chemical technologies [2,3] and still is a subject of recent mathematical inquiries [4]. One of the challenging problems is the appearance of chiral phases in achiral systems. Chiral symmetry breaking of this kind is responsible for many important processes in physics, chemistry, or biology and attracts great interest in modern science [5].

In solid-states physics chiral interactions also play a remarkable role, e.g., natural optical activity occurs owing to the intrinsic properties of materials that lack mirror symmetry. Magnetic chirality in crystals arises due to their crystallographic handedness. In magnetically ordered noncentrosymmetric crystals electronic spin-orbit scattering induces chiral asymmetry of exchange coupling, originating from quantum-mechanical Dzyaloshinsky-Moriya interactions [6]. Phenomenologically these are described by so called Lifshitz invariants, energy contributions linear in first spatial derivatives of the magnetization $\mathbf{M}(\mathbf{r})$ [7]

$$M_i \frac{\partial M_j}{\partial x_l} - M_j \frac{\partial M_i}{\partial x_l} \tag{1}$$

(x_l is a spatial coordinate). These chiral interactions stabilize localized (vortices) and spatially modulated structures with a fixed rotation sense of the magnetization [7,8]. Such chiral modulated structures have been identified in a number of noncentrosymmetric ferromagnets, antiferromagnets, and alloys [9]. Magnetic vortices exist in the cubic chiral ferromagnet NiMn [10]. Symmetry breaking by stresses or applied magnetic or electric fields may induce chiral magnetic couplings also in centrosymmetric crystals [11]. For bulk magnetic materials such couplings are supposed to be very weak and effects due to them have not been observed experimentally.

The situation may radically change in small artificial structures such as ferromagnetic thin films, multilayers, nanowires, and nanodots. Within these nanomagnets, much stronger induced chiral couplings are expected due to large strains or high numbers of lattice imperfections. Additionally, the broken symmetry at surfaces or interfaces is an important source for chiral effects in magnetic nanostructures [12–14]. While several mechanisms for chiral magnetic couplings due to symmetry breaking have been proposed [11,12,15], little is known about their influence on the magnetism of nanostructures. In this Letter we develop a phenomenological theory of chiral symmetry breaking and show that induced magnetic chirality may stabilize new magnetic structures and patterns in thin magnetic layers.

According to numerous experimental data on ferromagnetic layer systems, inhomogeneous stresses created by lattice mismatch, related defects, or interdiffusion between magnetic and nonmagnetic layers substantially influence their magnetic properties [16]. Under these conditions, induced chiral couplings should be inhomogeneous within magnetic nanostructures. Thus, the phenomenological chiral energy density can be written as

$$w_D = D\,\eta(\mathbf{r})L(\mathbf{m})\,. \tag{2}$$

Here D is a constant, L is a Lifshitz invariant of type (1), and a pseudoscalar function $\eta(\mathbf{r})$ describes the inhomogeneous distribution of the magnetic chiral energy. This function $\eta(\mathbf{r})$ plays the role of a chiral order parameter and may be treated as a physical field additional to the magnetization field which we take as unity vectors $\mathbf{m}(\mathbf{r}) = \mathbf{M}(\mathbf{r})/M_0$ ($M_0 = |\mathbf{M}|$). Possible distributions of the order parameter $\eta(\mathbf{r})$ can be described by a Landau-Ginsburg–type interaction functional with the density

$$w_c = \sum_i \tilde{A}\left(\frac{\partial \eta}{\partial x_i}\right)^2 + f(\eta^2) + D\,\eta(\mathbf{r})L(\mathbf{m})\,. \tag{3}$$

The density (3) includes the coupling with magnetization (2), a stiffness energy with constant $\tilde{A}$, and a homogeneous energy contribution $f(\eta^2)$. The magnetic energy density may be expressed by

$$w_m = A\sum_i \left(\frac{\partial \mathbf{m}}{\partial x_i}\right)^2 + w_a - \mathbf{m}\cdot\mathbf{h} - \frac{1}{2}\,\mathbf{m}\cdot\mathbf{h}_d\,, \tag{4}$$

which consists of exchange interaction with a stiffness constant A, magnetic anisotropy w_a, energy of the interaction

with an external $\mathbf{h} = \mathbf{H}/M_0$, and a demagnetizing field $\mathbf{h}_d = \mathbf{H}_d/M_0$. To obtain the equilibrium configurations of $\mathbf{m}(\mathbf{r})$ and $\eta(\mathbf{r})$ within the magnetic nanostructure, one has to find the minimum of the energy functional

$$W = \int \tilde{w}[\mathbf{m}(\mathbf{r}), \eta(\mathbf{r})]\, d\mathbf{r}\,, \tag{5}$$

where $\tilde{w} = w_c + w_m$, taking into account the equations of magnetostatics for the demagnetizing field $\mathbf{h}_d$. Surface chiral interactions are created by specific mechanisms. Thus, the related order parameter on surfaces will have certain fixed values $\eta(\mathbf{r})|_S = \eta_i^0$ which should be used as boundary conditions for the variational problem. In general η_i^0 may even vary within the surfaces.

The general features of the theory are demonstrated for the important case of magnetic films or layers sandwiched between nonmagnetic materials. This is modeled by a magnetic plate infinite in x and y directions confined by parallel planar surfaces at $z = \pm l$. We consider an (induced) uniaxial magnetic anisotropy $w_a = -Km_z^2$ much stronger than any higher order anisotropies responsible for orientational effects in the basal plane of the films. The parameter η has fixed values on the surfaces $\eta_{z=l} = \eta_1^0$, $\eta_{z=-l} = \eta_2^0$ and varies along the z direction.

For $K < 0$ the magnetization vector lies in the xoy plane (easy-plane type of the magnetization) and does not create demagnetizing fields on the surfaces. In this case, the Lifshitz invariant responsible for chiral effects can be written as

$$L = m_x \frac{dm_y}{dz} - m_y \frac{dm_x}{dz}\,. \tag{6}$$

The rotation of the vector $\mathbf{m}$ in the basal plane is described by an angle φ. The energy density in (5) depends only on the z coordinate and can be written as

$$\tilde{w} = A\left(\frac{d\varphi}{dz}\right)^2 + D\eta\,\frac{d\varphi}{dz} + \tilde{A}\left(\frac{d\eta}{dz}\right)^2 + f(\eta^2) - \kappa\cos(\eta\varphi)\,, \tag{7}$$

where the last term describes an in-plane anisotropy with $n = 2, 4, 6, \ldots$ depending on lattice symmetry and/or homogeneous strain. For this case, physically meaningful solutions from minimization of the functional (5) are found subject to the boundary conditions $d\varphi/dz(\pm l) = -D/(2A)\eta(\pm l)$. For $\kappa \neq 0$, the corresponding Euler equations must be solved numerically. For $\kappa \equiv 0$, the solutions are $\Delta\varphi = -D(2A)^{-1}\int_{-l}^{z} d\xi\,\eta(\xi)$ and $z + c_1 = \tilde{A}^{1/2}\int d\eta\,[f(\eta^2) - D^2\eta^2(4A)^{-1} + c_2]^{-1/2}$, where constants c_1, c_2 are determined by the boundary conditions for η. Typical solutions for $f(\eta^2) = a\eta^2 + b\eta^4$ are shown in Fig. 1. They represent inhomogeneous structures even in the case of zero volume chirality $[a > D^2/(4A), b > 0]$. Then chirally distorted magnetization is induced only by the boundary conditions, and the rotation has the highest values near the surfaces and slows into the volume of the layer [see Figs. 1(b) and 1(d)]. Under the influence of the surface chirality the order parameter η has a finite value even in the absence of the volume contribution. In the case of a finite volume chirality $[a < D^2/(4A)]$, modulated structures with a different rotation sense arise. An in-plane anisotropy $\kappa \neq 0$ may yield drastically reduced magnetization rotation [Figs. 1(c) and 1(d)]. Only a sizable volume chirality may overcome this anisotropy threshold and cause magnetization rotation also in the film volume [see insets in Fig. 1(d)].

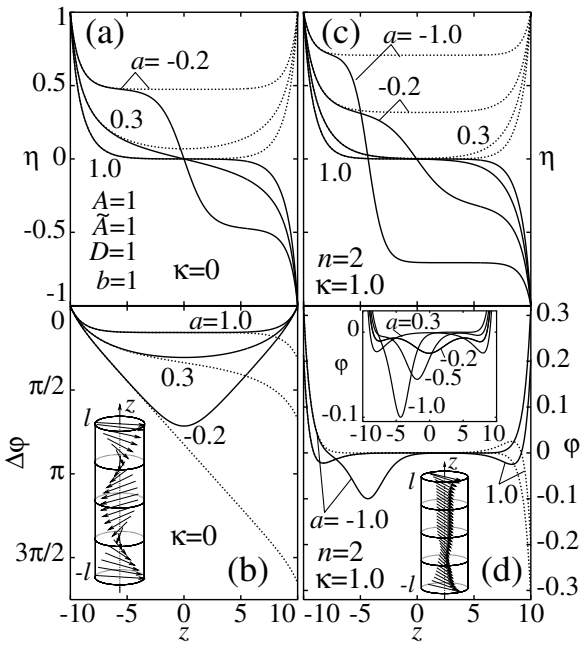

FIG. 1. Easy-plane ferromagnetic film: induced chiral modulations perpendicular to the film plane (z direction). Solutions for functional Eq. (7) with boundary conditions $\eta(\pm l) = \pm 1$ (full lines in all parts of the figure) or for $\eta(\pm l) = 1$ (dotted lines). (a) Chirality field without in-plane magnetic anisotropy displays the effect of decreasing a, i.e., growing strength of volume chirality. (b) Corresponding azimuth angle φ of the magnetization [inset: magnetization distribution in the film for $a = -0.2$ and $\eta(\pm l) = \pm 1$]. (c) Chirality field as in (a) but with in-plane anisotropy. (d) Corresponding φ. Inserted diagram is a magnification for the film volume [inset: magnetization distribution for $a = -1.0$ and $\eta(\pm l) = \pm 1$].

In layers with perpendicular anisotropy ($K > 0$), chiral effects are due to Lifshitz invariants with gradients along x and y directions. One of the possible invariants of this type is [8]

$$L = \left(m_z\frac{\partial m_x}{\partial x} - m_x\frac{\partial m_z}{\partial x} + m_z\frac{\partial m_y}{\partial y} - m_y\frac{\partial m_z}{\partial y}\right). \tag{8}$$

In this case, modulated structures are stable only if η is sufficiently large [of order $(AK)^{1/2}/D$]. For smaller values of η the uniaxial anisotropy suppresses modulated phases pinning the magnetization along the z axis. We consider possible localized chiral structures in this practically important case. It turns out that a weak chiral field $\eta(z)$ stabilizes two-dimensional states with finite extension, so called magnetic vortices, within the uniformly magnetized

matrix. Introducing spherical coordinates for the vector $\mathbf{m} = (\sin\theta\cos\psi, \sin\theta\sin\psi, \cos\theta)$ and cylindrical coordinates for the spatial variable $\mathbf{r} = (\rho\cos\zeta, \rho\sin\zeta, z)$, one finds that the variational problem has axisymmetric localized solutions $\psi = \zeta$, $\theta = \theta(\rho, z)$ with $\theta(0) = \pi$ and $\theta(\infty) = 0$ (Fig. 2). The equation for the internal stray field has an exact analytical solution and this part of the stray field energy may be incorporated into the uniaxial anisotropy. Here, we suppose that the constant K is sufficiently large to ignore the surface stray fields. In any case, the demagnetization influence of the layer surfaces gives an additional stabilization effect on the localized structures. The equilibrium values of $\eta(z)$ and $\theta(\rho, z)$ are determined by variation of the functional with the density

$$\tilde{w} = \left\{\tilde{A}\left(\frac{d\eta}{dz}\right)^2 + f(\eta^2) + D\eta\left(\frac{\partial\theta}{\partial\rho} + \frac{\sin\theta\cos\theta}{\rho}\right) + A\left[\left(\frac{\partial\theta}{\partial z}\right)^2 + \left(\frac{\partial\theta}{\partial\rho}\right)^2 + \frac{\sin^2\theta}{\rho^2}\right] - K\cos^2\theta\right\}\rho\,. \tag{9}$$

Because the magnetic layer is mainly homogeneously magnetized the coupling with the magnetization field practically does not influence the distribution of the $\eta(z)$ in this case. The equilibrium function $\eta(z)$ may be calculated independently from the magnetic subsystem, and the Euler equation for $\theta(\rho, z)$,

$$A\left(\frac{\partial^2\theta}{\partial z^2} + \frac{\partial^2\theta}{\partial\rho^2} + \frac{1}{\rho}\frac{\partial\theta}{\partial\rho} - \frac{\sin\theta\cos\theta}{\rho^2}\right) - D\eta(z)\frac{\sin^2\theta}{\rho} - K\sin\theta\cos\theta = 0\,, \tag{10}$$

includes the chiral parameter as a definite function of z which describes the distribution of the chiral order parameter under the influence of the surface chirality. Equation (10) was solved numerically for a number of functions $f(\eta^2)$. Solutions with well defined sizes exist for arbitrarily small values of η. In particular, for $f = B\eta^2$ ($B > 0$) (zero volume chirality) with symmetric boundary conditions $\eta_{z=l} = \eta_{z=-l} = \eta_1$, we have $\eta(z) = \eta_1\cosh(kz)/\cosh(kl)$ [with $k = (B/\tilde{A})^{1/2}$]. The corresponding solutions of Eq. (10) describe a magnetic vortex with increasing localization into the depth of the layer (Figs. 2 and 3). For these structures, the chiral interactions are decisive to stabilize these magnetic vortices which, otherwise, spontaneously collapse under the influence of magnetic fields or anisotropy [8].

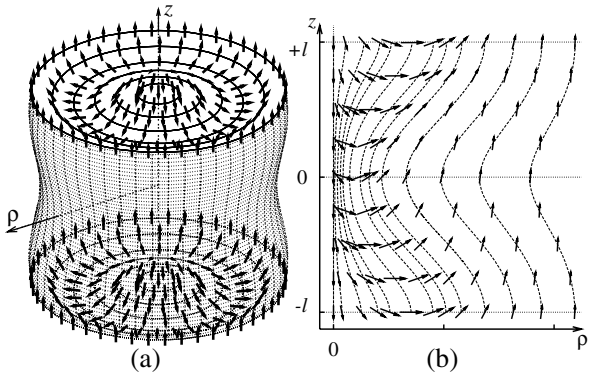

FIG. 2. Magnetization structure (depicted by arrows) of a vortex induced by chiral interactions in an inhomogeneous perpendicular ferromagnetic film. (a) View on top of the film. (b) Cut in the ρz half-plane. Dotted lines are curves for $\theta(\rho, z) = \text{const}$.

Interactions of type (1) are based on orbital magnetism [6]. An order of magnitude per bond near surfaces can be estimated by the ratio between isotropic exchange J and the Dzyaloshinsky-Moriya constant J_D, $J_D/J \simeq \Delta g/g \simeq \mu_L/\mu_S$, where g is the gyromagnetic ratio, Δg is its deviation from the free-electron value, and μ_L and μ_S are orbital and spin moments, respectively [6]. Within a tight-binding approximation [13], or for indirect exchange mechanisms [12,15], estimates $J_D/J \simeq 0.1$ were found for bonds which do not have a center of inversion at their midpoint. For example, each nearest-neighbor bond in a (001) surface of a fcc crystal contributes an antisymmetric exchange coupling [cf. Fig. 2(c) in Ref. [13]], which is equivalent to the Lifshitz invariant (8). Similarly, reduced symmetry near surface steps may induce further strong antisymmetric couplings [14]. Such considerations yield surface energy densities η_0, i.e., boundary conditions for η, with an order of magnitude $0.1\times$ isotropic exchange energy density. Equation (3) embodies a microscopic length describing a finite thickness of the surface inhomogeneity. This length should be in the thickness range where strong changes in orbital magnetism are observed in magnetic layers. Recent experimental data show considerably enhanced ratios of μ_L/μ_S in magnetic layers compared to the corresponding bulk materials [17]. Layer-resolved methods reveal variations of orbital moments into the depth of magnetic layers [18]. These experimental findings underpin our assumptions about strong inhomogeneous induced

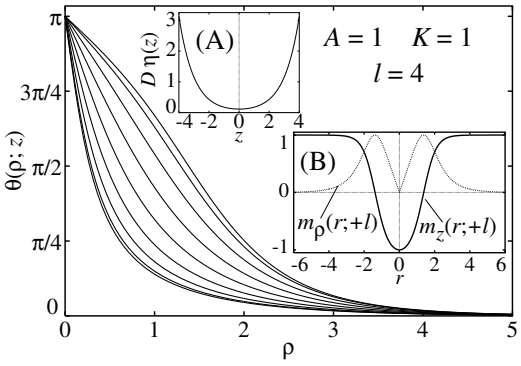

FIG. 3. Example of an induced magnetic vortex in a film with thickness $2l$. The main figure shows profiles for the magnetization tilt angle $\theta(\rho; z)$, solution of Eq. (10), for $z = ih/8$ ($i = 0, \ldots, 8$), i.e., from film midplane $z = 0$ (lowest curve) to film surface $z = l$ (top curve). Inset (A): fixed, inhomogeneous chirality in the film. Inset (B): vertical and radial magnetization component at the film surface along a line through the vortex center at $r = 0$.

chiral interactions in nanomagnets. The characteristic size of chiral structures Λ_0 (period of a modulated state or the size of the vortex core) is determined by the value of induced orbital moment. It can be estimated by $\Lambda_0 \sim A/D \sim 10Ja_0^2/(J_D a_0) \sim 10a_0(\mu_S/\mu_L)$, where a_0 is the lattice constant.

There are various experimental reports on chiral effects in magnetic layers; however, systematic searches for such effects and their causes were not yet reported and the relation to surface-induced chirality is generally ignored. Induced chiral exchange may be responsible (together with induced uniaxial anisotropy), e.g., for inhomogeneous surface states as recently discovered for $FeBO_3$ [19], for "chiral magnetic domains" observed in ultrathin FePd films [20], and for anomalous magnetic domains in layered perovskite manganites [21]. Helical structures induced in magnetic layers and thin films by tensile stresses are described in [22]. Twisted structures in layers with in-plane magnetization (Fig. 1) should become observable with the methods of Refs. [17,18]. Magnetic vortices (Figs. 2 and 3) may exist only in perpendicular magnetized films. For example, Ni/Cu(001) would be a convenient system with perpendicular magnetization in a broad range of thickness and low in-plane anisotropy. Locally focused, perpendicular magnetic field pulses could be used to nucleate vortices and to probe their stability in such films. We note that imperfections of the layer may stabilize chiral magnetic vortices. As an experimental confirmation, we refer to a recent observation of free magnetic vortices in thin buckled layers of permalloy [23]. We suggest that these vortices are due to uniaxial anisotropy and chiral interactions induced by large strain effects in this case.

We have shown that surface-induced chiral symmetry breaking in magnetic nanostructures should have a strong impact on their properties. It may cause chiral spatially modulated or localized magnetic structures which were previously associated only with low-symmetry crystals [24].

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