Helical phase in two-dimensional magnets due to four-spin interactions

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Abstract. We demonstrate that in ferromagnets with the D\textsubscript{3h} point group of symmetry a possible origin of phase transition from a collinear ferromagnetic state to a non-collinear state can be the fourth order contributions to the free energy density that are allowed by this point group of symmetry. At the same time, Dzyaloshinskii-Moria interaction vanishes in such materials. Via symmetry analysis we derive seven possible fourth order contributions to the free energy density with respect to the unit vector of the local magnetization direction but only two of them can be considered as independent. Moreover, for two-dimensional systems only one survives. Considered symmetry class is essential because a large group of two-dimensional intrinsic ferromagnets belongs to it, for example a monolayer Fe\textsubscript{3}GeTe\textsubscript{2}. The four-spin chiral exchange does also manifest itself in peculiar magnon spectra and favors spin waves.

1. Introduction
The discovery of single-layer graphene in 2004 attracted intense interest in the field of two-dimensional (2D) materials. The field has been growing ever since and large number of other stable atomically thin crystals have been studied, such as hexagonal hBN, MoS\textsubscript{2} and other dichalcogenides and layered oxides. Potential applications of monolayers include spin-based computer logic and new ways to store information that require possibility to stabilize and effectively manipulate magnetic order [1]. In particular, noncollinear magnetic structures, as skyrmions, might become the basis for future memory devices [2].

However, 2D magnets have not been realized in experiments for a long time. Early attempts mainly focused on inducing magnetism in 2D nonmagnetic materials by magnetic defects, including vacancies, doping impurities, localized electronic states arising at the edges [3-4]. But the implementation of long-range magnetic order in 2D materials is limited by these methods. Another method for observing magnetism in 2D materials is the effect of magnetic proximity in heterostructures. For example, it was shown in [5] that in the graphene/ferromagnet heterostructure, it is possible to effectively modulate spin current in the graphene layer by changing magnetization of the
ferromagnetic thin film. This do 2D magnetism possible, but the complex structure and fabrication at nanoscale severely limit its application [1]. Only in 2017 a long-range magnetic order was discovered in 2D van der Waals materials Cr$_2$Ge$_2$Te$_6$ [6] and CrI$_3$ [7]. Also, intrinsic ferromagnetism was described in atomic thin Fe$_2$GeTe$_2$ [8, 9]. Moreover, stable chiral magnetic textures have been observed in two different heterostructures based on Fe$_2$GeTe$_2$ [10, 11] and in thin layers of this material [12]. It is known that noncollinear magnetic order requires the existence of an antisymmetric exchange interaction called the Dzyaloshinsky-Moria interaction (DMI) [13, 14]. However, the origin of noncollinear spin states in Fe$_2$GeTe$_2$ cannot be correctly explained by DMI [15,16].

In this paper we discuss another possible origin of non-collinear states in ferromagnets with the D$_{3h}$ group symmetry, for which all LI terms in micromagnetic functional are forbidden, while multi-spin exchange interactions are allowed by symmetry [17]. We propose that four-spin chiral exchange interaction in such 2D magnets causes a phase transition from a collinear state to a non-collinear ground state. The paper is presented based on the results of the article [19].

2. Helical phase in magnet with D3h symmetry
Using standard symmetry analysis [18], we identify seven fourth order contributions to the free energy density that are allowed by D$_{3h}$ point group of symmetry. Only two invariants out of seven can be chosen as independent. Moreover, for 2D structures we are left with a single fourth order term. So, the free energy functional of ferromagnet with D$_{3h}$ point group of symmetry reads:

$$
F[m] = \int d^2r \left[ A \sum_{\alpha} (\nabla m_\alpha)^2 + 8Bm_\nu(m_\nu^2 - 3m_\nu^2)[\nabla \times m]_\nu + Km_\nu^2 - Hm \right],
$$

where $H$ stands for external magnetic field measured in energy unit, $A > 0$, $K$ – anisotropy constants. The first term represents usual symmetric exchange, the second term describes independent fourth order invariant which corresponds to four-spin chiral interaction.

Next, we consider a non-collinear ansatz for local magnetization field $m(r)$ that can be written in a form:

$$
m(r) = n \cos \alpha + [m_1 \cos(kr) + m_2 \sin(kr)] \sin \alpha,
$$

where $n$, $m_1$ and $m_2$ are mutually perpendicular unit vectors: $m_1 = (\sin \phi, -\cos \phi, 0)^T$, $m_2 = (\cos \phi \cos \theta, \sin \phi \cos \theta, -\sin \theta)^T$, $n = m_1 \times m_2$. The angle $\alpha$ corresponds to the transition from collinear state, $\alpha = 0$, to a pure helix state, $\alpha = \pi/2$.

We substitute this ansatz (2) into (1) and minimize with respect to wave vector and find the free energy as:

$$
\frac{F[m]}{VB^2} = \frac{9}{64} (\sin \alpha + 5 \sin 3 \alpha) \sin^4 \theta \cos^2 \theta + \frac{KA}{B^2} \cos^2 \theta \cos^2 \alpha + \frac{KA}{2B^2} \sin^2 \theta \sin^2 \alpha - Hm.
$$

In the absence of the external field the free energy is parametrized by a single dimensionless parameter $KA/B^2$. The figure 1 provides the dependence of an angle describing a transfer from collinear to helix state ($\alpha$) on both constant of magnetic anisotropy and external magnetic field parallel to the plane ($H_z=H_y=0$). It is seen the sharp phase transitions from the collinear to the helical phase and back with K increasing. The corresponding jumps are also seen in the left panel. The maximum achieved value of $\alpha$ is $\alpha = \pi/6$. Naturally for large value of anisotropy or an external magnetic field, their effect suppresses the effect of four-spin interaction and the stable state is collinear. The effect of the four-spin interactions is thus in some way similar to the DMI.

3. Magnon spectra
Let us now investigate how the four-spin chiral interaction may affect the magnon spectra. To develop the magnon dispersion it is needed to solve the Landau-Lifshits equation:

$$
\frac{dm}{dt} = m \times H_{eff},
$$

where $H_{eff} = -\delta F/\delta m$ with respect to a small variation $\delta m$. 


We consider collinear ground state which is characterized by a constant magnetization vector $\mathbf{n}$. For the case of the external magnetic field is $\mathbf{H} = H_0(\sin \theta \cos \varphi, \sin \theta \sin \varphi, \cos \theta)$ $\mathbf{n}$ reads:

$$\mathbf{n} = \left( \frac{\lambda \sin \theta \cos \varphi}{1 - \frac{K \lambda}{H_0}}, \frac{\lambda \sin \theta \sin \varphi}{1 - \frac{K \lambda}{H_0}}, \frac{\lambda \cos \theta}{1 - \frac{K \lambda}{H_0}} \right),$$

(5)

where $\lambda = f(K/H_0, \theta)$ is the solution of the equation $|\mathbf{n}| = 1$ that corresponds to the minimal free energy. It is equal to 1 in the absence of anisotropy, $\lambda = f(0, \theta) = 1$.

**Figure 1.** Phase diagram of the collinear and non-collinear phases as a function of anisotropy and external magnetic field in lateral direction. The left panel shows three vertical cross sections for different constants of anisotropy.

Using the ansatz $\mathbf{m}(\mathbf{r}, t) = \mathbf{n} + (\mathbf{n} \times \delta)e^{i\mathbf{q}\mathbf{r} - i\omega t}$, we linearize the Landau-Lifshits equation with respect to $\delta$ and find the magnon dispersion:

$$\omega_q^\pm = \pm S(q^2, \lambda) + \frac{3\lambda^3 B_q}{1 - \frac{K \lambda}{H_0}} \sin^2 \theta \cos \theta \sin(3\varphi + \chi),$$

(7)

where $\chi$ is the angle between the direction of propagation of magnon and the direction of in-plane component of magnetic field. In the case of zero anisotropy, $S$ is just $S|_{K=0} = Aq^2 + H_0$. We notice the symmetry $\omega_q^+ = -\omega_q^-$. The maximal asymmetry $\omega_q^+ - \omega_q^-$ is reached when $\chi = \pm \pi/2 - 2\varphi$. Moreover, the asymmetry is linear with respect to $B_q$ and has non-trivial dependence on $\theta$, the polar angle of the external magnetic field (see figure 2).

**Figure 2.** Dependence of the asymmetry on the magnetic field polar angle for different values of anisotropy strength $K/H_0$. 

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4. Conclusion
In conclusion, we demonstrate that fourth order contribution $\omega(r) = 8Bm_y(m_x^2 - 3m_y^2)(\nabla \times m)_z$ to the free energy density is responsible for the appearance of a long-range non-collinear magnetic order in ferromagnets with $D_{3h}$ group of symmetry. It looks compatible with spin spirals observed in a recent experiment on Fe$_3$GeTe$_2$. Also, we showed how such four-spin chiral interaction in 2D magnets leads to non-reciprocal magnon dispersion.

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