Asymmetric dispersion relation in spin-spiral structures

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The asymmetric spin-wave dispersion relation observed recently [Zakeri *et al.*, Phys. Rev. Lett. **104**, 137203 (2010)], is explained within a quantum model consisting of an anisotropic Heisenberg coupling and the Dzyaloshinski-Moriya interaction (DMI). Applying a transformation of the spin operators into a representation without fixed quantization axis, a Green's function technique for finite temperatures allows to obtain the spin-wave dispersion for various magnetic spiral structures including cycloidal and conical ones. In case of vanishing anisotropy, the DMI drives an incommensurate antiferromagnetic alignment forming a cycloidal spin spiral. The spin-wave dispersion is symmetric. For nonzero anisotropy, the spins are aligned in a transverse conical spiral and the dispersion becomes asymmetric in agreement with the experimental observation. The asymmetry is reduced with increasing temperature.

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There is a considerable effort in understanding complex magnetic structures.[1] The high resolution of spin-polarized scanning tunneling microscopy allows the detection of more complicated magnetic patterns such as in a Mn monolayer on W(110) and W(100) surfaces.[2–4] Ultrathin Fe film grown on W(110) is another system where a very refined magnetic structure had been observed likewise.[5–7]

Theoretically a spiral alignment of the spins can arise from a competition between nearest neighbors and next-nearest neighbors. However, such an explanation does not reflect the breaking of inversion symmetry in thin films. Otherwise, it is well established that the relativistic Dzyaloshinski-Moriya interaction (DMI) plays an important role in systems without inversion symmetry.[8,9] Phenomenologically, the antisymmetric DMI between two adjacent magnetic atoms reads $E_{12}=\mathbf{D}_{12}\cdot(\mathbf{S}_1\times\mathbf{S}_2)$. The coupling vector $\mathbf{D}_{12}$ is antisymmetric and disappears in case of an inversion symmetry between the lattice sites 1 and 2. Originally the DMI had been introduced[8] to explain the weak ferromagnetism in antiferromagnetic material, for instance, in $MnCO_3$ or CrF_3 compounds. Due to the competition between the isotropic Heisenberg antiferromagnetic coupling and the DMI, a canted magnetic structure is realized leading to an uncompensated weak magnetization. The role of the DMI is widely discussed in a broad class of materials such as frustrated systems,[10] the induced spin chirality in spin chains,[11] cubic magnets at low temperatures,[12] entanglement of the Ising model with DMI,[13] and valence-bond systems.[14] Notice that a comprehensive effective theory for helimagnets is discussed in Ref. 15. The study of various spin alignments is raised additionally by the search for new types of order in magnetoelectric multiferroics in which both magnetic and ferroelectric order coexist.[16–18] A characteristic property of magnetic multiferroics is the occurrence of an electric polarization due to a spiral magnetic ordering.[19–21] The discovery of multiferroics was accompanied by a theoretical progress in the description of such noncollinear structures on a microscopic scale[22,23] and a mesoscopic one.[24] The results are regarded as an inverse effect of the DMI. Multiferroic perovskites are analyzed in Ref. 25 and moreover, the relationship between ferroelectricity and DMI is discussed in Ref. 26 as well as an exchange bias driven by DMI in antiferromagnetic interfaces.[27] Recently the dynamical interplay between ferroelectricity and magnetism is analyzed including the DMI.[28]

The seminal studies of multiferroics has initiated the present one which is focused on the recently observed asymmetric spin-wave dispersion relation on Fe(110).[29] In Ref. 29, the first experimental evidence is given of the influence of the DMI on the spin-wave dispersion in an Fe double layer grown on W(110). The experimental results are in agreement with theoretical predictions based on a classical spin-wave model.[30] Otherwise the approach[30] yields no prediction for the concrete realization of the spiral states favored by the DMI and furthermore, the results are obtained for fixed temperatures. Very recently, the significance of temperature effects on the magnon spectrum has been pointed out.[31] From a microscopic point of view, the quantum DMI modifies the spectrum of the excitation energy. It is therefore the aim of the present Brief Report to elucidate the relation between the spin-wave excitation energy and the magnetic alignment of the spins. The progress can be reached by applying a representation of the underlying spin operators with an arbitrary quantization axis.[32] This approach enables us to include a broad class of spin-spiral structures. The Hamiltonian includes both, the isotropic Heisenberg coupling, an anisotropic interaction and the DMI. The spin-wave energy is found by calculating the temperature-dependent retarded Green's function matrix. Minimizing the ground-state energy yields the direction of the quantization axis. The method allows also to find out the temperature dependence of the spin-wave excitations and its asymmetry.

An extended Heisenberg model is chosen to represent the underlying magnetic system. The isotropic Heisenberg model H_1 describes the coupling of the spins in the magnetic system and drives the parallel or antiparallel alignment. The asymmetric DMI H_2 favors a canting of the spins in the plane perpendicular to the DM vector $\mathbf{D}$. Because of the asymmetric superexchange interaction of the magnetic moments, the spins orient in a noncollinear fashion, which can be the origin of a spiral-spin structure of several types. The Hamiltonian reads

$$H = H_1 + H_2 + H_3, \tag{1}$$

where the three part are defined by

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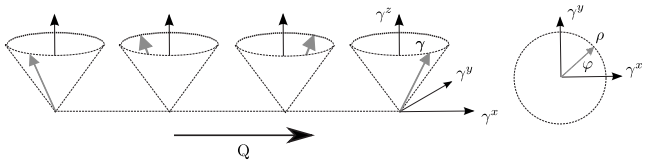

FIG. 1. Quantization axis of transformed operators with $\rho = \sqrt{1-(\gamma^\tau)^2}$.

$$H_1 = -\frac{1}{2}\sum_{ij} J_{ij}\mathbf{S}_i\mathbf{S}_j - g\mu_B\mathbf{H}^\mathbf{A}\sum_i \mathbf{S}_i, \tag{2}$$

$$H_2 = -\frac{1}{2}\sum_{ij} \mathbf{D}_{ij}(\mathbf{S}_i \times \mathbf{S}_j),$$

$$H_3 = -\frac{1}{2}\sum_{ij} J_{ij}^A S_i^\tau S_j^\tau. \tag{3}$$

An anisotropy in a certain τ direction is included by the third term H_3. In the upcoming calculations, we will restricted ourself to $\tau=z$. In H_1, the summation is taken over all nearest neighbors and J_{ij} denotes the symmetric exchange interaction. The spin couples also to an intrinsic anisotropy field $\mathbf{H}^\mathbf{A}$ establishing a spiral structure out off the x-y plane provided this field points in z direction. The relativistic spin-orbit interaction reflected in the DMI H_2 is determined by the antisymmetric coupling $\mathbf{D}_{ij}$.

For the description of magnetic systems with a spiral structure, some special cases had been discussed in Refs. 33 and 34, it is appropriate to transform the spin operators to the eigenrepresentation of the quantization axis. Here, the quantization axis, see Fig. 1, at site f of the lattice is given by the real unit vector $\gamma_f=(\gamma_f^x,\gamma_f^y,\gamma_f^z)=(\rho\cos\mathbf{Q}\mathbf{r}_f,\rho\sin\mathbf{Q}\mathbf{r}_f,\gamma_f^z)$. The respective transformation of the spin-$\frac{1}{2}$ operators with $\sigma_f=\frac{1}{2}-b_f^+b_f$ is given in terms of Pauli operators b^+ and b. The most general form is

$$S_f^\alpha = \gamma_f^\alpha\sigma_f + A_f^\alpha b_f + (A_f^*)^\alpha b_f^+. \tag{4}$$

The coefficients in Eq. (4) are chosen in such a manner that the commutation relations of spin operators are guaranteed. Using the realization[32] $A_f^x=-\frac{1}{2}[\gamma^z\cos\varphi_f+i\sin\varphi_f]$, $A_f^y=-\frac{1}{2}[\gamma^z\sin\varphi_f-i\cos\varphi_f]$, $A_f^z=\frac{\rho}{2}$ the Hamiltonian is transformed into in the following compressed form where only even terms in the annihilation and creation operators has to be taken into account:

$$H = -\frac{1}{2}\sum_{ij}\{X_{ij}\sigma_i\sigma_j + 2S_{ij}b_ib_j^+ + R_{ij}b_ib_j + \tilde{R}_{ij}b_i^+b_j^+\} - g\mu_B\sum_i (H^A)^\alpha\gamma_i^\alpha\sigma_i. \tag{5}$$

The prefactors in the Hamiltonian represent the renormalized interaction between the spins including the Heisenberg interaction and the asymmetric DMI pointing in z direction. The Fourier-transformed prefactors are defined in the subsequent section. Notice that within this representation, the asymmetry is reflected by the relation $S_{ij}\neq S_{ji}$.

In many-particle systems, the thermodynamic Green's function method is a powerful tool to elucidate the spin-wave dispersion relation given by the poles of the Green's function defined as $\langle\langle b(t);b^+(t')\rangle\rangle=-i\Theta(t-t')\langle[b(t),b^+(t')]_-\rangle$. Here, the temperature dependence enters by the ensemble average $\langle\cdots\rangle$. The decoupling of the equations of motion is done by random phase approximation. After some algebra, the excitation energy of spin waves can be derived by the eigenvalues of the following matrix equation

$$\underbrace{\begin{pmatrix}\omega-\varepsilon_1(\mathbf{q},\mathbf{Q}) & \varepsilon_2(\mathbf{q},\mathbf{Q})\\ -\varepsilon_2(\mathbf{q},\mathbf{Q}) & \omega+\varepsilon_1(-\mathbf{q},\mathbf{Q})\end{pmatrix}}_{\Lambda}\begin{pmatrix}\langle\langle b;b^+\rangle\rangle_\mathbf{q} & \langle\langle b;b\rangle\rangle_\mathbf{q}\\ \langle\langle b^+;b^+\rangle\rangle_\mathbf{q} & \langle\langle b^+;b\rangle\rangle_\mathbf{q}\end{pmatrix} = \begin{pmatrix}2\langle\sigma\rangle & 0\\ 0 & -2\langle\sigma\rangle\end{pmatrix}, \tag{6}$$

where $\varepsilon_1(\mathbf{q},\mathbf{Q})=\langle\sigma\rangle[X(0,\mathbf{Q})-2S(\mathbf{q},\mathbf{Q})]+g\mu_B(H^A)^\alpha\gamma^\alpha$ and $\varepsilon_2(\mathbf{q},\mathbf{Q})=2\langle\sigma\rangle R(\mathbf{q},\mathbf{Q})=\langle\sigma\rangle\frac{\rho^2}{2}[J(\mathbf{q})+J^A(\mathbf{q})-\frac{1}{2}P(\mathbf{q},\mathbf{Q})]$ with $X(\mathbf{q},\mathbf{Q})=[J(\mathbf{q})+J^A(\mathbf{q})](\gamma^z)^2-\frac{\rho^2}{2}P(\mathbf{q},\mathbf{Q})$ and $S(\mathbf{q},\mathbf{Q})=\frac{1}{4}[J(\mathbf{q})+J^A(\mathbf{q})]\rho^2-\frac{1}{8}[(\gamma^z)^2+1]P(\mathbf{q},\mathbf{Q})+\frac{1}{4}\gamma^zV(\mathbf{q},\mathbf{Q})$. The magnetization points in direction of the magnetic anisotropy. Spin waves of wave vector $\mathbf{q}$ and the direction of the propagation of the incommensurate spin spiral with wave vector $\mathbf{Q}$ are defined by the quantization axis $\boldsymbol{\gamma}$. In case of $\tau=z$, the spiral orientations vary around the z direction. The deviation from the collinear state is characterized by $\gamma^\tau<1$. In the parallel aligned state, the condition reads $\gamma^\tau=1$. The cycloidal or screw state corresponds to $\gamma^\tau=0$. The wave vector of the spin spiral is determined by minimizing the free energy. The dispersion relation of the low-lying states reads

$$\varepsilon(\mathbf{q},\mathbf{Q}) = \frac{\gamma^\tau\langle\sigma\rangle}{2}V(\mathbf{q},\mathbf{Q}) \pm \sqrt{\frac{1}{2}\left\{(\gamma^\tau\langle\sigma\rangle)^2\left[J(\mathbf{q})+J^A(\mathbf{q})-\frac{1}{2}P(\mathbf{q},\mathbf{Q})\right]-\langle\sigma\rangle^2\left[J(\mathbf{q})+J^A(\mathbf{q})-\frac{1}{2}P(0,\mathbf{Q})\right]\right\}[P(0,\mathbf{Q})-P(\mathbf{q},\mathbf{Q})]} \tag{7}$$

with

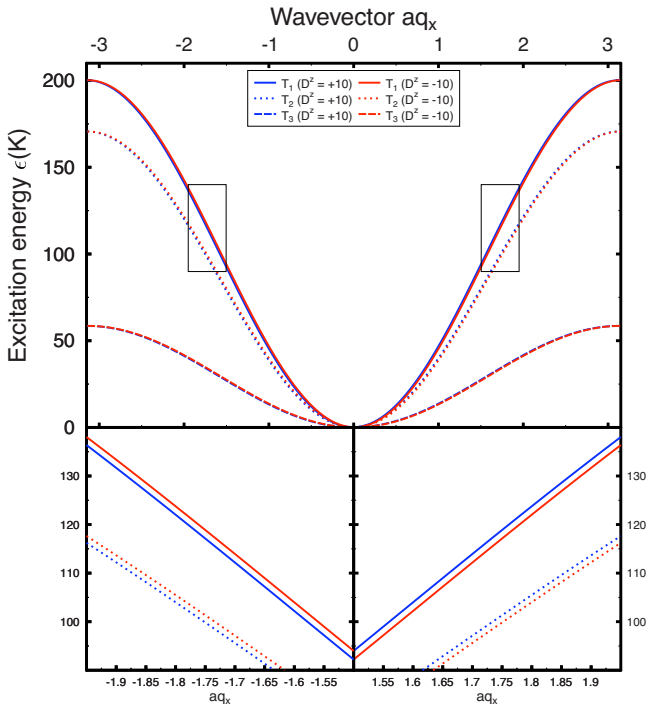

FIG. 2. (Color online) Asymmetric spin-wave dispersion relation for two different chirality (red/blue), at three different temperatures $T_1=1$ K, $T_2=50$ K and $T_3=95$ K. The lower graphs show the change in the excitation energy around $aq_x=\frac{\pi}{2}$, if the chirality is reversed. The phase transition temperature is about $T_c=100$ K. The parameters read $J=100$ K, $J^A=0.1$ K, $|D^z|=10$ K, $H^A=0.0001$ K, and $\gamma^z=0.6$.

$$\gamma^\tau\langle\sigma\rangle = \frac{g\mu_B H^A}{\frac{1}{2}P(0,\mathbf{Q}) - [J(0) + J^A(0)]}. \tag{8}$$

The last equation determines the wave vector $\mathbf{Q}$. The chirality is defined by the rotational sense of this vector. The dispersion relation of the magnetic system obtained in Eq. (7) consists of two parts. The asymmetric first part, the function $V(\mathbf{q},\mathbf{Q})$ is defined below in Eq. (9), only contributes in case of a spin alignment with an additional component in the direction of the DM vector. This can be interpreted as a conical spin spiral. The second part is symmetric with a renormalized interaction $P(\mathbf{q},\mathbf{Q})=J(\mathbf{q}+\mathbf{Q})+J(\mathbf{q}-\mathbf{Q})+i[D^\tau(\mathbf{q}-\mathbf{Q})-D^\tau(\mathbf{q}+\mathbf{Q})]$ and contributes also in case of a screw or cycloidal spin ordering. The positive branch of the spin-wave dispersion relation is chosen because a negative excitation energy indicates an instability of the system. In case of $\gamma^\tau\neq 0$, the dispersion relation becomes asymmetric. The rotational sense (chirality) is defined by the interplay between the different interactions of the magnetic system. This results in a wave vector $\mathbf{Q}$ related to a constant canting of spins between adjacent lattice sites. The rotational sense depends on the sign of the DMI, allowing only one type of chirality, which is in accordance with Ref. 2. In Fig. 2, the calculated spin-wave spectrum is shown along q_x.

The asymmetry is characterized by

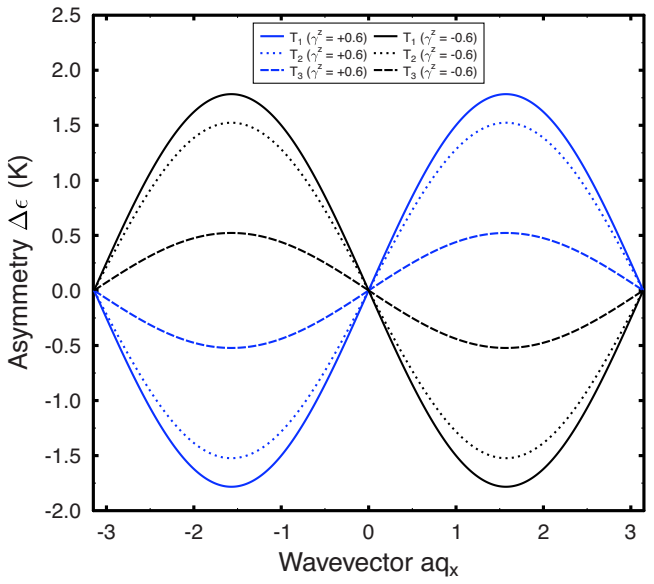

FIG. 3. (Color online) Asymmetry of energy as a function of wave vector for two different directions of magnetization at three different temperatures ($T_1<T_2<T_3$). For a fixed temperature, this behavior is observed in Ref. 29.

$$\Delta\varepsilon(\mathbf{q},\mathbf{Q}) = \gamma^\tau\langle\sigma\rangle V(\mathbf{q},\mathbf{Q}) = \gamma^\tau\langle\sigma\rangle\{J(\mathbf{q}+\mathbf{Q}) - J(\mathbf{q}-\mathbf{Q}) + i[D^\tau(\mathbf{q}+\mathbf{Q}) + D^\tau(\mathbf{q}-\mathbf{Q})]\}. \tag{9}$$

Here both, the symmetric J_{ij} and the antisymmetric interaction D_{ij}^τ contribute to the chirality-dependent asymmetry. In accordance with recent calculations,[30] only the components of the DM vector parallel to the ground-state magnetization γ^τ influence the dispersion relation. In case of all moments are perpendicular to the anisotropy direction ($\gamma^\tau=0$) the asymmetry vanishes. The asymmetry is depicted in Fig. 3. It shows a distinct maximum at $aq_x=\frac{\pi}{2}$, with the lattice constant a. A switching of the magnetization is equivalent to a change in sign of γ^τ and affects the asymmetry in agreement with recent experimental results.[29] The deviations of the asymmetry in experiments to those of the model are assumed to be related to the restriction to nearest-neighbor interaction and a different coordination number. Because the Green's function, defined in Eq. (6), includes the statistical average we find also the temperature dependence of the asymmetry via the temperature dependence of the magnetization $\langle\sigma\rangle$. With increasing temperature, the asymmetry is reduced as shown in Fig. 3.

Magnetic systems of spin $\frac{1}{2}$ with noncollinear spin structures are investigated in the framework of temperature-dependent two-time retarded Green's function technique. The inclusion of the asymmetric Dzyaloshinskii-Moriya interaction leads to an asymmetric spin-wave dispersion relation in case of conical spirals. The induced asymmetry in the energy is governed by the symmetric Heisenberg exchange coupling, the anisotropy, and the antisymmetric DMI. The chirality is represented by the incommensurate wave vector of the spin spiral. The method enables the inclusion of

temperature effects. Here, a reduction in the asymmetry is observed for increasing temperature. A cycloidal or screw *alignment of the spins would not lead to an asymmetric dispersion relation. Referring to recent experimental results ruling out longitudinal conical spin spirals, the magnetic ordering in these systems has to be of transverse conical type. The asymmetry is calculated for a cubic lattice with nearest-neighbor interaction. Although not so pronounced, compared to thin films, the asymmetry is also expected to occur in multiferroic spinel oxids.

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