Asymmetric Magnon Excitation by Spontaneous Toroidal Ordering

Satoru Hayami\textsuperscript{1,2,*}, Hiroaki Kusunose\textsuperscript{3}, and Yukitoshi Motome\textsuperscript{2}

\textsuperscript{1}Theoretical Division, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA
\textsuperscript{2}Department of Applied Physics, University of Tokyo, Tokyo 113-8656, Japan
\textsuperscript{3}Department of Physics, Meiji University, Kanagawa 214-8571, Japan

Effects of spontaneous toroidal ordering on magnetic excitation are theoretically investigated for a localized spin model that includes a staggered Dzyaloshinsky-Moriya interaction and anisotropic exchange interactions, which arise from the antisymmetric spin-orbit coupling and the multi-orbital correlation effect. We show that the model exhibits a Néel-type antiferromagnetic order, which simultaneously accompanies a ferroic toroidal order. We find that the occurrence of toroidal order modulates the magnon dispersion in an asymmetric way with respect to the wave number: a toroidal dipole order on the zigzag chain leads to a band-bottom shift, while a toroidal octupole order on the honeycomb lattice gives rise to a valley splitting. These asymmetric magnon excitations could be a source of unusual magnetic responses, such as nonreciprocal magnon transport. A variety of modulations are discussed while changing the lattice and magnetic symmetries. Implications of candidate materials for asymmetric magnon excitations are presented.

Fig. 1. (Color online) Schematic pictures of the zigzag chain along the $x$ direction for (a) the localized spin model in Eq. (1) and (b) the itinerant electron model in Eq. (2). (c) Schematic picture of the honeycomb lattice for the localized spin model in the $xy$ plane. In (a) and (c), $A$ and $B$ represent two sublattices.
tion of the centrosymmetric lattices with local asymmetry. We consider the Heisenberg model supplemented by anisotropic exchange interactions and a sublattice-dependent DM interaction, whose Hamiltonian is given by

$$\mathcal{H} = \sum_{i,j} t_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + \sum_{i,j} G_{ij} \left( \mathbf{S}_i^z \mathbf{S}_j^z - \mathbf{S}_i^+ \mathbf{S}_j^- - \mathbf{S}_i^- \mathbf{S}_j^+ \right) + D \sum_p \left[ (\mathbf{S}_{Ap} \times \mathbf{S}_{Ap+1}) - (\mathbf{S}_{Bp} \times \mathbf{S}_{Bp+1}) \right] \cdot \mathbf{z},$$

(1)

where $\mathbf{S}_i$ is the $S = 1/2$ operator at site $i = (l, p)$ ($l$ and $p$ denote the indices for the sublattice A and B, and unit cell, respectively). The first term represents the Heisenberg-type exchange coupling for nearest-neighbor (NN) and next-nearest-neighbor (NNN) spins, $J_1$ and $J_2$, respectively. The second term in Eq. (1) is the anisotropic exchange interactions for NN and NNN spins, $G_1$ and $G_2$, respectively. The former $G_1$ originates from the multi-orbital correlation effect, while the latter $G_2$ from the ASOC, as discussed below. The third term in Eq. (1) is the antisymmetric exchange interaction between NNN spins (DM interaction)\textsuperscript{12-14} [see Fig. 1(a)], whose origin is also the ASOC as shown below. Note that there is no DM interaction for NN spins because of the inversion center at the bond median.

In the second and third terms in Eq. (1), the sublattice-dependent ASOC inherited from the local asymmetry of the zigzag lattice plays an important role. This is understood, for instance, by considering the strong-coupling limit of an effective single-band model,\textsuperscript{5} whose Hamiltonian is given by

$$\mathcal{H} = \sum_{i,\sigma} t_{i\sigma} (c_{i\sigma}^\dagger c_{i\sigma} + \text{H.c.}) + U \sum_{i} n_{i\uparrow} n_{i\downarrow} + \sum_{i,\sigma,\sigma'} g(k) \cdot \mathbf{\sigma}_{\sigma\sigma'} (c_{i\sigma}^\dagger c_{i\sigma'}^\dagger - c_{i\sigma'} c_{i\sigma}) \cdot (\mathbf{e}_{\text{ff}} \times \mathbf{e}_{\text{ff}}),$$

(2)

The first and second terms comprise the Hubbard model in a standard notation; $t_1$ and $t_2$ are NN and NNN hoppings, respectively. The third term represents the sublattice-dependent ASOC in the form of the spin-dependent electron hopping, which originates from the atomic SOC, local asymmetry, and multi-orbital hybridization effects between orbitals with different parity.\textsuperscript{6,7,17} $g$ is the asymmetric vector with respect to $k$: $g(k) = 2\alpha \sin(ka)\mathbf{z}$ [$\alpha$ is the amplitude, $a$ is the lattice constant ($a = 1$), and $\mathbf{z}$ is the unit vector along the $z$ axis; see Figs. 1(a) and 1(b)], and $\mathbf{\sigma}$ is the Pauli matrix. The sign factor comes from the zigzag structure, as an electron at the A and B sublattice sites feels an opposite potential gradient. By considering the strong-coupling limit of the model in Eq. (2) at half-filling, we obtain the coupling constants in the model in Eq. (1) by the second-order perturbation in terms of $t_m/U$ ($m = 1, 2$) and $a/U$ as $J_m = 4t_m^2/U$, $G_1 = 4a^2/U$, and $D = 2 \sqrt{J_2 G_2}$. On the other hand, in general, the multi-orbital correlation effect, which is not taken into account in Eq. (2), favors the Néel-type AFM ordering along the $z$ direction in the presence of the atomic SOC and the multi-orbital hybridization, as demonstrated in a $d$-p model\textsuperscript{18} and an extended Kondo lattice model.\textsuperscript{9} Such a multi-orbital correlation effect is incorporated in the $G_1$ term in Eq. (1). Hereafter, we consider $J_1 > 0$ and $J_2 \geq 0$ with taking $J_1$ as an energy unit, and $G_1 \geq 0$, $G_2 \geq 0$, and $D = 2 \sqrt{J_2 G_2}$.

First, we examine the ground state of the model in Eq. (1). To stabilize the long-range orders, we here consider a three-dimensional system composed of ferromagnetically weakly-coupled 1D zigzag chain. We obtain the ground-state phase diagram by the Luttinger-Tisza method by treating the spins as classical vectors.\textsuperscript{19} The results are plotted in Figs. 2(a) and 2(b) for $J_2 = 0$ and $J_2 = 0.2$, respectively. The contour displays the optimal wave number $q^*$. When $J_2 = G_1 = G_2 = 0$ [the origin in Fig. 2(a)], the stable spin configuration is a $q = 0$ collinear AFM without any spin anisotropy. With increasing $G_1$ ($G_2$), the NN (NNN) anisotropic interaction stabilizes the AFM ordering along the $z$ direction (z-AFM) [in the $xy$ plane (x-AFM)] without any spin canting. Note that the system has the spin-rotational symmetry around the $z$ axis. With further increasing $G_1$ and $G_2$, incommensurate magnetic orders with longer period $q \neq 0$ appear. As shown in Fig. 2(b), the $q \neq 0$ region becomes larger for larger $J_2$, as there is frustration between $J_1$ and $J_2$.

The results in Figs. 2(a) and 2(b) show that the model exhibits the collinear $z$-AFM order for sufficiently large $G_1/G_2$. As described above, the $z$-AFM order on the zigzag chain is of particular interest as it accompanies the odd-parity multipole, such as magnetic toroidal and magnetic quadrupoles, through the spin-orbit coupling effect when $D \neq 0$. The magnetic toroidal dipoles are we are considering in this study are defined by $t = \sum_i (r_i \times S_i)$, where $r_i$ is the position vector from the inversion center to the lattice site $i$. In such a situation, the z-AFM order possess the toroidal dipoles in the $x$ direction because $S_i \parallel z$ and $r_i \parallel y$ by taking the bond median as the origin.

Next, we examine magnetic excitations in the collinear $z$-AFM state by using the spin-wave theory. We adopt the standard Holstein-Primakoff transformation, which is represented by $S_{zq}^+ = \sqrt{2S} a_q^\dagger$ ($S_{zq}^- = \sqrt{2S} b_q^\dagger$), $S_{zq}^- = \sqrt{2S} a_q^\dagger$, and $S_{zq}^+ = S - a_q^\dagger a_q$ ($S_{zq}^- = -S + b_q^\dagger b_q$) for the A (B) sublattice. Here, $a$ and $b$ are the boson operators for the A and B sublattices, respectively, and $q$ is the wave number along the chain direction. We adopt the linear spin-wave approximation, in which the magnon-magnon interactions are ignored. Then, the spin-wave Hamiltonian in the $q$ space is obtained as

$$\mathcal{H} = E_0 + \sum_q (A_q^S + A_q^A)(a_q^\dagger a_q + b_q^\dagger b_q) + \sum_q B_q (a_q^\dagger b_q + a_q b_q^\dagger),$$

(3)
where $A_q^S = 2S [ (J_1 + G_1) + J_2(\cos q - 1) - G_2(\cos q + 1)]$, $A_q^{AS} = 2SD\sin q$, $B_q = 2S(J_1 - G_1)\cos(q/2)$, and $E_0 = -NS^2(J_1 - J_2 + G_1 - G_2)$. By using the Bogoliubov transformations $\alpha_q = a_q \cosh \theta_q + b_q^* \sinh \theta_q$ and $\beta_q^* = a_q \sinh \theta_q + b_q \cosh \theta_q$ [$\theta_q = (1/2) \tanh^{-1} (B_q/A_q^S)$], we can diagonalize Eq. (3) into the form of $\mathcal{H} = \sum_q \omega_q (\alpha_q^\dagger \alpha_q + \beta_q^\dagger \beta_q) + \text{const.}$, where the magnon dispersion relation $\omega_q$ is obtained as

$$\omega_q = \sqrt{(A_q^S)^2 - B_q^2} + A_q^{AS}. \quad (4)$$

Figure 3(a) shows the magnon dispersion in the $z$-AFM state at $J_2 = 0.1$, $G_1 = 0.05$, and $G_2 = 0.02$. The magnon excitation spectrum has a peculiar form: in addition to the gap opening, the dispersion undergoes an asymmetric deformation with respect to $q$, leading to a shift of the band bottom from $q = 0$. This is similar to the electronic band structure in the presence of the toroidal ordering. For comparison, we also show the results at $J_2 = G_1 = G_2 = 0$ (dotted lines) and $J_2 = G_2 = 0$ (dashed lines). The results indicate that $G_1$ opens the excitation gap and $D = 2\sqrt{JG_2}$ brings about the asymmetric deformation.

We analyze the effect of $G_1$, $G_2$, and $J_2$ more carefully. Figures 3(b) and 3(c) represent the band-bottom shift $\Delta q$ and the spin gap $\Delta s$ on the $J_2$-$G_2$ planes at $G_1 = 0.05$. The results show that $J_2$ and $G_2$ significantly affect the asymmetric deformation of the magnon dispersion, as shown in Fig. 3(b). This is because the deformation is caused by $A_q^{AS} \propto D\sin q$ originating from the sublattice-dependent DM interaction in the third term in Eq. (1). On the other hand, the spin gap depends on both $G_1$ and $G_2$, while it does not show strong $J_2$ dependence, except in the region close to an incommensurate phase, as shown in Fig. 3(c).

For further elucidating the relation between the magnon dispersion and the symmetry of the system, we extend our analysis to other cases. Namely, we consider both AFM and FM cases while changing the parameters in Eq. (1) beyond the range expected from the perturbation theory. We also consider a straight 1D chain in an applied electric field, which has a uniform DM interaction.

Figure 4 summarizes the characteristic of the magnon dispersions in the matrix form for the staggered/uniform DM interactions (row) and the AFM/FM orders (column). The above result for the spontaneous $z$-AFM ordering on the zigzag chain is shown in the upper-left panel. A similar asymmetric dispersion also appears for the $z$-FM ordering under the uniform DM interaction (lower-right). Note that the latter is in the same category as the toroidal magnon discussed for the helical magnetic structure in an applied magnetic field. These asymmetric magnon dispersions lead to peculiar magneto-optical phenomena, such as the nonreciprocal directional dichroism.

On the other hand, when the $z$-AFM order occurs on the straight chain with the uniform DM, the magnon dispersion undergoes a different modulation (lower-left), which is similar to a “Rashba-type” splitting in the electronic band structure under a uniform ASOC. Meanwhile, in the case of the $z$-FM order on the zigzag chain, there is a symmetric modulation, reflecting the spatial inversion symmetry (upper-right).

Finally, let us extend the analysis to the two-dimensional case. We study the magnon dispersion in the collinear $z$-AFM state on the honeycomb lattice. Similar to the 1D zigzag case, the spin model includes the anisotropic and antisymmetric interactions, as shown in Fig. 1(c). $D$ is positive (negative) in the counterclockwise (clockwise) direction in the hexagonal plaquettes. In the case of the honeycomb lattice, a collinear $z$-AFM order accompanies ferroic toroidal octupoles instead of dipoles, reflecting the presence of threefold rotational sym-
metry of the lattice, i.e., the net toroidal-dipole component is zero. We find that, by the Luttinger-Tisza analysis, such a $z$-AFM order with toroidal octupoles becomes stable in the ground state for large $G_1/G_2$, although the region becomes narrower than that for the zigzag chain, as shown in Fig. 5(a).

In the collinear $z$-AFM ordered state, the magnon dispersion does not show any shift of the band bottom, as the lowest contribution to the ASOC is in the third order in $(q_x, q_y)$. Note that the band-bottom shift is obtained also on the honeycomb lattice once a uniaxial pressure is applied, since it breaks the threefold rotational symmetry. Instead, the asymmetry appears near the Brillouin zone boundary around the $K$ and $K'$ points, as shown in Fig. 5(b). This is a valley splitting, similar to the electronic band structure discussed in noncentrosymmetric compounds, such as monolayer transition metal dichalcogenides.\textsuperscript{27} Note that the band-bottom shift is obtained also on the honeycomb lattice once a uniaxial pressure is applied, since it breaks the threefold rotational symmetry and induces a $q$-linear contribution to the ASOC as in the zigzag chain case.

To summarize, we have clarified that spontaneous ordering of toroidal multipoles modulates the magnon excitations in an asymmetric way in the momentum space. The observation of asymmetric magnon spectra will provide an experimental probe for toroidal multipoles. There are many experimental candidates. One is the AFM zigzag compound $\alpha$-Cu$_2$V$_2$O$_7$.\textsuperscript{28} Since the compound may possess both the staggered and uniform DM interactions due to the peculiar magnetic and lattice structures, both a band-bottom shift and “Rashba-type” splitting are expected to be observed. Another candidates are transition metal tricoganoides, such as MnPS$_3$ and MnPSe$_3$;\textsuperscript{29,31} the $z$-AFM order on the honeycomb lattice will lead to a valley splitting in the magnon spectrum. A distorted honeycomb compound $\beta$-YbAlB$_4$ is also a candidate, as it shows the AFM ordering under a uniaxial pressure.\textsuperscript{32} A band-bottom shift is expected in addition to a valley splitting. We note that the diamond-lattice systems will also be in the present scope, as they possess the similar physics related with the local asymmetry.

Similar asymmetric excitations will be obtained also in itinerant electron systems, such as the Hubbard and Kondo lattice models, when the sublattice-dependent ASOC is present. Reflecting the itinerant electron degree of freedom, asymmetric Stoner-type excitations are expected in addition to asymmetric magnon excitations. Such an analysis is left for future study.

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