Orbital Angular Momentum of Magnons in Collinear Magnets

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We study the orbital angular momentum of magnons for collinear ferromagnet (FM) and antiferromagnetic (AF) systems with nontrivial networks of exchange interactions. The orbital angular momentum of magnons for AF and FM zig-zag and honeycomb lattices becomes nonzero when the lattice contains two inequivalent sites and is largest at the avoided-crossing points or extremum of the frequency bands. Hence, the arrangement of exchange interactions may play a more important role at producing the orbital angular momentum of magnons than the spin-orbit coupling energy and the resulting non-collinear arrangement of spins.

For more than a century, scientists have been intrigued by the conversion of spin into orbital angular momentum (OAM) and vice versa. In 1915, A. Einstein and W.J. de Haas [1] demonstrated that a change of magnetization can cause the container of that magnet to rotate. Also in 1915, S.J. Barnett [2] demonstrated that the rotation of electrons can be converted into magnetization. In solids, the conversion of spin into orbital angular momentum is produced by the spin-orbit (SO) coupling. Recently, scientists have been searching for evidence of OAM [3, 4] in spin excitations, also known as magnons. Whereas a magnon corresponding to a single spin flip has spin $S = \pm \hbar$, the OAM $\mathcal{L}$ of such a magnon is unknown.

Two main approaches have been employed to search for the OAM of magnons. Because SO coupling is also responsible for Dzyaloshinskii-Moriya (DM) interactions, Neumann et al. [5] examined the OAM of magnons associated with the non-collinear spin states produced by DM interactions. Other groups have investigated the OAM of magnons in confined geometries. In a whispering gallery mode cavity, for example, circulating magnons with perpendicular OAM can be excited on the surface of a FM sphere by incident light [6–8]. Magnons with a range of perpendicular OAM can be excited on the surface of a FM magnon band topology.

We study the orbital angular momentum of magnons for collinear ferromagnet (FM) and antiferromagnet (AF) systems with nontrivial networks of exchange interactions. The orbital angular momentum of magnons for AF and FM zig-zag and honeycomb lattices becomes nonzero when the lattice contains two inequivalent sites and is largest at the avoided-crossing points or extremum of the magnon bands. For FM zig-zag chains, the OAM vanishes when the upper and lower bands cross but becomes quite large when the gap between the bands is small but nonzero. For FM honeycomb lattices, the upper and lower bands carry opposite OAM when averaged over the Brillouin zone (BZ). For AF honeycomb lattices, the two degenerate magnon bands can be divided into major and minor branches that carry different OAM. We shall see that the OAM and Berry curvature [5] capture different aspects of the magnon band topology.

Formally, the classical equations of motion [1, 2] for the dynamical magnetization $\mathbf{\mu}_i = 2\mu_B \delta \mathbf{S}_i$ at site $i$ produce the linear momentum $p_{i\alpha}$ [3]:

$$p_{i\alpha} = \frac{1}{4\mu_B M_0} (\mathbf{\mu}_i \times \mathbf{n}_i) \cdot \frac{\partial \mathbf{\mu}_i}{\partial x_\alpha},$$

where $M_0 = 2\mu_BS$ is the static magnetization for a spin $\mathbf{S}_i$ pointing along $\mathbf{n}_i$ (a derivation of the classical OAM is provided in the Supplementary Material [18]). Using the $1/S$ quantization conditions $\pi_i^+ = \mu_{ix} n_{iz} + i \mu_{iy}$ and $\pi_i^- = \mu_{ix} n_{iz} - i \mu_{iy} = 2\mu_B \sqrt{2S} \hat{a}_i^\dagger$ for the dynamical magnetization in terms of the local Boson operators $a_i$ and $a_i^\dagger$ satisfying the momentum-space commutation relations \{\hat{a}_k^{(r)}, \hat{a}_k^{(s)^\dagger}\} = \delta_{rs} \delta_{k,k'}$ and $\{a_k^{(r)}, \hat{a}_k^{(s)^\dagger}\} = a_k^{(r)}, a_k^{(s)^\dagger} = 0$, the quantized OAM along $\mathbf{z}$ is given by

$$\mathcal{L}_z = \sum_i (r_i \times p_i) \cdot \mathbf{z} = \frac{\hbar}{2} \sum_{r,s=1}^{M} \sum_k \left\{ \hat{a}_k^{(r)} \hat{a}_k^{(s)^\dagger} - \hat{a}_k^{(s)^\dagger} \hat{a}_k^{(r)} \right\},$$

where $r$ and $s$ refer to the $M$ sites in the magnetic unit cell and

$$\hat{a}_k^{(r)} = -i \left( k_x \frac{\partial}{\partial k_y} - k_y \frac{\partial}{\partial k_x} \right)$$

$$\hat{a}_k^{(s)^\dagger} = i \left( k_x \frac{\partial}{\partial k_y} + k_y \frac{\partial}{\partial k_x} \right)$$

This Letter demonstrates that collinear magnets with tailored exchange geometries can generate magnons that exhibit OAM. Results for both FM and AF zig-zag and honeycomb lattices in two dimensions indicate that the OAM becomes nonzero when the lattice contains two inequivalent sites and is greatest at the avoided-crossing points or extremum of the magnon bands. For FM zig-zag chains, the OAM vanishes when the upper and lower bands cross but becomes quite large when the gap between the bands is small but nonzero. For FM honeycomb lattices, the upper and lower bands carry opposite OAM when averaged over the Brillouin zone (BZ). For AF honeycomb lattices, the two degenerate magnon bands can be divided into major and minor branches that carry different OAM. We shall see that the OAM and Berry curvature [5] capture different aspects of the magnon band topology.
we must diagonalize
\[ \exp(\mathbf{H}) \]
the Hamiltonian \( J \) of the matrix \( a_q \), and the dashed line shows \( k_y a / 2 \pi = 0.1 \). (d) Magnons of an \( r = 2 \) FM zig-zag material traveling with opposite momenta \( \pm \mathbf{k} \) and OAM \( \pm \mathbf{L} \) in a temperature gradient.

is the OAM operator. Transforming to the Boson operators \( b_r^{(n)} \) and \( b_r^{(n)\dagger} \) that diagonalize the Hamiltonian \( H \), we define \[ a^{(r)} = \sum_n \left( X^{-1}(k)_{rn} b_r^{(n)} + X^{-1}(k)_{rn+M} b_{r+M}^{(n)} \right), \] (4)
\[ a^{(r)\dagger} = \sum_n \left( X^{-1}(k)_{r+n} b_r^{(n)} + X^{-1}(k)_{r+n+M} b_{r+M}^{(n)} \right). \]

The zero-temperature expectation value of \( \mathcal{L}_z \) for magnon state \( b_k^{(n)\dagger}|0 \rangle = |k, n \rangle \) with frequency \( \omega_n(k) \) is
\[ \mathcal{L}_z(k) = \langle k, n | \mathcal{L}_z | k, n \rangle = \frac{\hbar}{2} \sum_{r=1}^M \left\{ X^{-1}(k)_{rn} \hat{L}_{k} X^{-1}(k)^*_{rn} - X^{-1}(k)_{r+m,n} \hat{L}_{k} X^{-1}(k)^*_{r+m,n} \right\}. \] (5)

For collinear spin states without DM interactions, \( X^{-1}(-k) = X^{-1}(k)^* \) so that \( \mathcal{L}_z(k) = -\mathcal{L}_z(-k) \) is an odd function of \( k \).

i. FM zig zag. Our first case study is the square lattice shown in Fig. 1(a) with alternating FM bonds \( J_1 > 0 \) and \( J_2 > 0 \) coupling sites 1 and 2 with spins up. Second order in the operator \( \mathbf{v}_k = (a^{(1)}_k, a^{(2)}_k, a^{(1)\dagger}_k, a^{(2)\dagger}_k) \), the Hamiltonian \( H_2 = \sum_k \mathbf{v}_k \cdot \hat{L}(k) \cdot \mathbf{v}_k \) is defined in terms of the matrix
\[ \hat{L}(k) = (J_1 + J_2) S \begin{pmatrix} 1 & -\Psi_k^* & 0 & 0 \\ -\Psi_k & 1 & 0 & 0 \\ 0 & 0 & 1 & -\Psi_k^* \\ 0 & 0 & -\Psi_k & 1 \end{pmatrix}, \] (6)
where \( \Psi_k = (J_1 \xi_k + J_2 \xi_k) / (J_1 + J_2) \) with \( \xi_k = \exp(i k_x a) + \exp(i k_y a) \). To study the magnon dynamics, we must diagonalize \( \hat{L} \cdot \mathbf{N} \), where
\[ \mathbf{N} = \begin{pmatrix} I & 0 \\ 0 & -I \end{pmatrix} \] (7)
and \( I \) is the two-dimensional identity matrix. Using the relation \( \mathbf{N} \cdot X(k) \cdot \mathbf{N} = X^{-1}(k) \) to normalize the eigenvectors \( | \Psi_k \rangle \), we find
\[ X^{-1}(k) = \frac{1}{\sqrt{2} \Psi_k} \begin{pmatrix} -\Psi_k^* & \Psi_k & 0 & 0 \\ 0 & |\Psi_k| & |\Psi_k| & 0 \\ 0 & 0 & |\Psi_k| & |\Psi_k| \end{pmatrix}. \] (8)

It is then simple to show that
\[ \mathcal{L}_z(k) = \frac{\hbar}{4 |\Psi_k|} \hat{L}(k) \cdot \mathbf{N}, \] (9)
and it is the same for magnon bands \( n = 1 \) and \( 2 \) with energies \( \hbar \omega_{1,2}(k) = (J_1 + J_2) S (1 \pm |\Psi_k|) \).

Results for \( \mathcal{L}_z(k) / \hbar \) are plotted as a function of \( r = J_2 / J_1 \) in Fig. 1(c) [20]. Not surprisingly, \( \mathcal{L}_z(k) \) vanishes for a square-lattice FM with \( r = 1 \). Comparing the “hot spots” in Fig. 1(c) for \( r = 1.1 \) with the magnon bands in Fig. 1(b) for \( k_y a / 2 \pi = 0.1 \), we see that the OAM is largest \( \sim 2 \hbar \) at the avoided-crossing points \( k^* \) of bands 1 and 2 near \( k_x a / 2 \pi = 0.4 \). As \( r \) increases, the gap between the bands grows, the region of large positive and negative \( \mathcal{L}_z(k) \) spreads out in \( k \) space, and its amplitude decreases. For very large \( r \), the regions of large positive and negative \( \mathcal{L}_z(k) \) stretch into stripes. The wavevectors \( k^* \) are associated with a sign change in the Berry curvature [5, 18].

ii. AF zig zag. For the square lattice in Fig. 2(a), we take \( J_1 < 0 \) and \( J_2 > 0 \) so that sites 1 and 2 have spins up while sites 3 and 4 have spins down. Although \( \hat{L}(k) \) is 8 dimensional, it breaks into the two identical 4 \times 4 matrices
\[ \hat{L}(k)' = (J_2 - J_1) S \begin{pmatrix} 1 & -\gamma_1 \xi_k & 0 & \gamma_2 \xi_k \\ -\gamma_2 \xi_k & 1 & \gamma_1 \xi_k & 0 \\ 0 & \gamma_1 \xi_k & 1 & -\gamma_2 \xi_k \\ \gamma_1 \xi_k & 0 & -\gamma_2 \xi_k & 1 \end{pmatrix}. \] (10)
with doubly degenerate magnon energies

\[ \hbar \omega_{1,2}(k) = 2(J - J_1)S \left\{ 1 - (\gamma_1^2 - \gamma_2^2)|\xi_k|^2 \right\} \]

\[ \pm \gamma_2 \sqrt{\gamma_1^2(\xi_k^2 - \xi_n^2)^2 + 4|\xi_k|^2} \right\}^{1/2}, \] (11)

where \( \gamma_n = J_n/2(J_2 - J_1) \).

While no simple analytic expression for the OAM is possible, we readily obtain the numerical solutions in Fig. 2(b). For \( J_1 = 0 \), the zig-zag chains are isolated from each other and the numerical solution is identical to one for FM zig-zag chains. Hence, the two bands have the same OAM. When \( J_1 = -J_2 \), the lower band exhibits a larger amplitude of the OAM than the upper band, as seen in the central panel of Fig. 2(b). When \( J_2 = 0.01 \) and \( J_1 = -1 \), the FM interaction within each zig-zag chain is very weak while the AF interaction between chains is strong. Then the OAM is only significant around discrete points \( k^* \) along the line \( k_x = k_y \). As expected, the OAM vanishes as \( J_2/J_1 \to 0 \).

iii. FM honeycomb. We now consider the honeycomb lattice shown in Fig. 3(a) with FM exchange coupling \( J > 0 \). Provided that the easy-axis anisotropy \(-K \sum_i S_{iz}^2\) is sufficiently strong, we may also add a DM interaction \( D \) between next-neighbor sites without tilting the spins. We then find

\[ L(k) = \frac{3JS}{2} \begin{pmatrix} 1-G_k & -\Gamma_k & 0 & 0 \\ -\Gamma_k & 1+G_k & 0 & 0 \\ 0 & 0 & 1+G_k & -\Gamma_k^* \\ 0 & 0 & -\Gamma_k^* & 1-G_k \end{pmatrix}, \] (12)

where \( G_k = d \Theta_k \) with \( d = -2D/3J \), \( \Theta_k = 4 \cos(3k_xa/2) \sin(\sqrt{3}k_ya/2) - 2 \sin(\sqrt{3}k_ya) \), and

\[ \Gamma_k = \frac{1}{3} \left\{ e^{ik_xa} + e^{-i(k_x+\sqrt{3}k_y)a/2} + e^{-i(k_x-\sqrt{3}k_y)a/2} \right\}. \] (13)

Because the anisotropy \( \kappa = K/J \) merely shifts the magnon energies \( \hbar \omega_{1,2}(k) = 3JS(1 - \kappa \pm g_k) \) with \( g_k = \sqrt{\Gamma_k^2 + G_k^2} \) but does not affect the OAM, we neglect its contribution to \( L(k) \). After the usual manipulations, we find \( X^{-1}(k)_{11} = -1/2c_1g_k \), \( X^{-1}(k)_{12} = 1/2c_2g_k \), \( X^{-1}(k)_{21} = (G_k + g_k)/2c_1\Gamma_kg_k \), and \( X^{-1}(k)_{22} = -(G_k - g_k)/2c_2\Gamma_kg_k \), where \( 1/c_1^2 = 2g_k(g_k + G_k) \) and \( 1/c_2^2 = 2g_k(g_k + G_k) \). The 31, 32, 41, and 42 matrix elements of \( X^{-1}(k) \) vanish.

For \( d = 0 \), the upper and lower band frequencies \( \omega_1(k) \) and \( \omega_2(k) \) cross at \( k^* = (1/3, \sqrt{3}/3, 2\pi/a) \) and equivalent points throughout the BZ. With

\[ L_{2n}(k) = \frac{\hbar}{4|\Gamma_k|} \begin{pmatrix} \Gamma_k^* \\ \Gamma_k^* \end{pmatrix}, \] (14)

the OAM is the same for both bands. Notice that this expression is the same as Eq. (9) for \( L_{2n}(k) \) of the FM zig-zag lattice with \( \Psi_k \) replaced by \( \Gamma_k \). As seen in Fig. 3(b), \( L_{2n}(k)/\hbar \) has modest values of \( \pm 3/16 = \pm 0.1875 \) at \( k^* \).

Since the DM interactions change sign upon spatial inversion, \( L_{2n}(k)/\hbar \) contains both even and odd terms with respect to \( k \) due to the \( G_k = -G_{-k} \sim d \) functions in \( X^{-1}(k) \). For \( d > 0 \), the averages of \( L_{2n}(k)/\hbar \) and \( L_{2z2}(k)/\hbar \) over the BZ are negative and positive, respectively. With increasing \( d \), a gap opens between the two magnon bands and \( |L_{2n}(k)| \) grows at the avoided-crossings points \( k^* \). For \( d = 0.01 \), the largest values of the OAM at \( k^* \) are about \( \pm 0.38\hbar \). The Berry curvature \( \kappa \) of the FM honeycomb lattice is discussed in the Supplementary Material [18].

iv. AF honeycomb. The final case study is the honeycomb lattice sketched in Fig. 4(a) with AF exchange \( J < 0 \) between alternating up and down spins. Since it shifts the magnon energies but does not affect the OAM, the DM interaction is neglected in the following discussion. We obtain

\[ L(k) = -\frac{3JS}{2} \begin{pmatrix} 1+\kappa & 0 & 0 & -\Gamma_k^* \\ 0 & 1+\kappa & -\Gamma_k & 0 \\ 0 & -\Gamma_k & 1+\kappa & 0 \\ -\Gamma_k^* & 0 & 0 & 1+\kappa \end{pmatrix}. \] (15)

The usual procedure yields \( X^{-1}(k)_{11} = -1/2c_1f_k \), \( X^{-1}(k)_{12} = 1/2c_2f_k \), \( X^{-1}(k)_{21} = (f_k + 1 + \kappa)/2c_1\Gamma_kf_k \), and \( X^{-1}(k)_{22} = (f_k - 1 - \kappa)/2c_1\Gamma_kf_k \), where \( 1/c_1^2 = \frac{\hbar^2}{4|\Gamma_k|^2} \).
2f_{k}(1 + \kappa + f_{k}) and 1/|c_{2}|^{2} = 2f_{k}(1 + \kappa - f_{k}) with f_{k} = \sqrt{(1 + \kappa)^{2} - |k|^2}$. Other matrix elements of $X^{-1}(k)_{rn}$ for modes $n = 1$ and 2 vanish.

Surprisingly, the doubly degenerate magnon bands with energies $\hbar \omega_{x,2}(k) = 3J/S\sqrt{(1 + \kappa)^{2} - |k|^{2}}$ exhibit distinct OAMs with

$$L_{z1}(k) = \frac{\hbar}{4} \left( 1 + \kappa + f_{k} \right) \frac{\Gamma_{k}}{|k|} \hat{z}_{k} \frac{\Gamma_{k}^{*}}{|k|},$$

and ratio $L_{z1}(k)/L_{z2}(k) = -(1 + \kappa + f_{k})/(1 + \kappa - f_{k}) < 0$. As seen in Fig. 4(b) for $\kappa = 0$, the major and minor bands have different patterns for $L_{zn}(k)$ but are both threefold symmetric. The maxima in $|L_{z1}(k)|/\hbar$ of 3/8 [21] appear at points $k_{*}$ where $\Gamma_{k}$ vanishes and $\hbar \omega_{n}(k)$ reaches a maximum of $3J/S$. Those points coincide with the avoided-crossing points $k_{*}$ of the non-degenerate bands for the FM honeycomb lattice.

For $\kappa > 0$, the average OAM $L_{av}(k) = (L_{z1}(k) + L_{z2}(k))/2$ of the major and minor bands of the AF honeycomb lattice equals the OAM of the $d = 0$ FM honeycomb lattice given by Eq. (14) and plotted in Fig. 3. We emphasize that the major and minor bands of the AF honeycomb lattice are identical in every other respect. For example, their spin-spin correlation functions $S_{\alpha\beta}(k, \omega)$ are equal [18].

The topological nature of quasiparticles in solids is often characterized by their Berry phase [5]. In momentum space, the Berry curvature is given by

$$\Omega_{n}(k) = \frac{i}{2\pi} \left\{ \nabla_{k} \times \langle u_{n}(k)|\nabla_{k}u_{n}(k) \rangle \right\} \cdot z,$$

where $|u_{n}(k)\rangle$ is the single-particle wave function of band $n$ and $\langle u_{n}(k)|\nabla_{k}u_{n}(k) \rangle$ is called the Berry connection. Integrating $\Omega_{n}(k)$ over the BZ then gives the Chern number $\mathcal{C}_{n}$. The connection between the Berry curvature and the OAM is clarified by rewriting Eq. (5) as

$$L_{zn}(k) = -i\hbar \left\{ k \times \langle u_{n}(k)|\nabla_{k}u_{n}(k) \rangle \right\} \cdot z.$$

Thus, while the Berry curvature is the curl of the Berry connection, the OAM is the cross product of the momentum $k$ and Berry connection.

At low energies and momenta, Eq. (19) reduces to the expression of Matsumoto and Murakami [11, 12] for FM magnons, which was parameterized in terms of an effective mass $m^{*}$. Since we are interested in the OAM of both FM and AF magnons throughout the BZ, we prefer using the more general expression given above. Because it is produced by SOC, the OAM discussed in Ref. [5] is not related to the one described by Eq. (19).

Theoretically, the OAM predicted in this paper vanishes for mode $n$ if the matrix elements $X^{-1}(k)_{rn}$ and $X^{-1}(k)_{r + M, n}$ can be simultaneously rotated onto the real axis by a suitable choice of normalization factor $c_{n}$ [18]. This generates terms like $\exp(ik_{x}a)$ and $\exp(ik_{y}a)$ that are not mixed with their complex conjugates in $L(k)$, $X^{-1}(k)_{rn}$, and $X^{-1}(k)_{r + M, n}$.

Whenever magnons exhibit OAM, the lattice contains two inequivalent sites either due to exchange (cases i and ii) or structure (cases iii and iv). In such a non-Bravais lattice, the violation of inversion symmetry about each site creates preferred channels for the magnons and an asymmetry in $k$ space that produces the OAM. In that sense, the present work follows in the spirit of earlier work on magnon confinement in spherical [6–8] and cylindrical [9, 10] geometries. We surmise that it may be easier to generate and control the OAM of magnons by designing devices with tailored exchange interactions than with customized SO couplings and spin textures.

In all four case studies, the largest OAM appears at the crossing points or extremum $k^{*}$ of the magnon bands. For the FM zig-zag lattice, a slight increase of $r = J_{2}/J_{1}$ from 1 has a huge effect on the OAM because it creates two inequivalent magnetic sites while opening a gap between the magnon bands at $k^{*}$. Increasing $r > 1$ further reduces the OAM while widening the gap between the magnon bands. Since the FM honeycomb lattice with $D = 0$ already contains two inequivalent sites, its magnons exhibit nonzero OAM at wavevectors $k^{*}$ and elsewhere throughout the BZ. By breaking the odd symmetry of $L_{zn}(k)$, a nonzero $D$ allows the upper and lower magnon bands to carry a net OAM when averaged over the BZ. Consequently, larger values of the OAM appear at $k^{*}$. Because it breaks the degeneracy of otherwise identical bands, the OAM of an AF honeycomb lattice is particularly intriguing.

This work opens the gateway for the future experimental study of the OAM of magnons in collinear spin systems. While bulk zig-zag systems with $J_{1} \approx J_{2} > 0$ (case i) are difficult to experimentally identify due to their similar exchange constants, many experimental systems can be described as zig zags coupled by AF exchange $J_{1} < 0$ (case ii). AF-coupled zig-zag chains decorate the quasi-two-dimensional honeycomb lattice compound $\text{Na}_{2}\text{CoO}_{2}\text{TeO}_{6}$ [22], the transition-metal phosphates $\text{XPS}_{3}$ ($\text{X} = \text{Fe or Ni}$) [23–25], and iridium-based compounds like $\text{Na}_{2}\text{IrO}_{3}$ [26]. Both the honeycomb sub-lattice of $\text{Li}_{2}\text{Ni}_{2}\text{SbO}_{6}$ [27] and the square AF sub-lattice of $\text{Ba}_{2}\text{Mn}(\text{PO}_{4})_{2}$ [28] also contain zig-zag chains. While many Ruddlesden-Popper manganites have zig-zag chains with AF correlations [29], the metallic manganite $\text{La}_{0.67}\text{Ca}_{0.33}\text{MnO}_{3}$ has zig-zag chains running within square AF $ab$-planes [30]. Due to their photoluminescent properties, many of these materials are candidates for opto-spintronics, which provides avenues to probe or perturb the OAM of magnons.

The magnetic phase diagrams of honeycomb systems with chemical formula $\text{ABX}_{3}$ were reviewed by Sivadas et al. [31]. Examples of FM honeycomb lattices (case iii) are $\text{CrSiTe}_{3}$ and $\text{CrGeTe}_{3}$ [32–34]. Another well-known Cr-based FM honeycomb system is $\text{CrI}_{3}$ [35], which has topological magnon excitations that were studied by Chen et al. [36]. AF honeycomb lattices (case iv)
are found in MnPS$_3$ and MnPSe$_3$ [37].

There are many physical consequences connected with the predicted OAM of magnons, including its effect on magnon decay rates, the scattering by photons and phonons, and the scattering of magnons in thermal gradients (see Fig. 1(d)) [38]. Once a magnon with momentum $k$ and OAM $L_{zn}(k)$ is created, conservation of total angular momentum $J = S + L$ (spin plus orbital) due to dipolar interactions has been demonstrated for small $ka$ [1, 2] even in the absence of SO coupling. Most present measurements of magnon transport do not probe the OAM $L_{zn}(k)$, which averages to zero over magnon bands within the BZ. While many issues remain to be explored, including the generalization of this work for non-collinear spin states, we have established that the magnons of two-dimensional collinear magnets can carry significant OAM provided that the exchange interactions meet some easily satisfied conditions. We hope that future theoretical and experimental work will explore the nature of that OAM and how it can be used to understand and control the properties of magnons in magnetic materials.

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CLASSICAL EQUATIONS OF MOTION, LINEAR MOMENTUM, AND OAM

We briefly review the classical equations of motion and Lagrangian formulation originally presented in Refs. [1, 2] for collinear spins. A general Hamiltonian can be written in terms of the magnetization $\mathbf{M}_i$ as

$$H = -\sum_{i=1}^{M} J_{\alpha \beta} M_{\alpha i} M_{\alpha i+1, \beta} - \sum_{i=1}^{M} \mathbf{M}_i \cdot \mathbf{h}_i - \frac{\kappa}{2} \sum_{i=1}^{M} M_{i}^2 - \frac{1}{8\pi} \sum_{i=1}^{M} h_i^2,$$  
(S1)

where we only consider one dimension for simplicity and the magnetic unit cell contains $M$ sites. Unless explicitly indicated, repeated Greek indices are summed. The magnetostatic equations for the magnetic dipole field $\mathbf{h}_i$ are $\nabla \times \mathbf{h}_i = 0$ and $\nabla \cdot \mathbf{h}_i = -4\pi \nabla \cdot \mathbf{M}_i$. The first can be satisfied by defining a scalar potential $\phi_i$ as $\mathbf{h}_i = \nabla \phi_i$. Asymmetric exchange interactions like the DM interaction may be included in $J_{\alpha \beta}$. It is also easy to include further-neighbor exchange interactions. The total magnetization $\mathbf{M}_i$ can be written in terms of the dynamical magnetization $\mathbf{\mu}_i$ as

$$\mathbf{M}_i = \mathbf{\mu}_i + n_i \sqrt{M_0^2 - \mathbf{\mu}_i^2},$$ (S2)

where the static magnetization $\mathbf{M}_0 = gS \mathbf{n}_i$ $(g = 2\mu_B)$ lies along $\mathbf{n}_i$ and $\mathbf{\mu}_i \cdot \mathbf{n}_i = 0$. Defining the effective field

$$\mathbf{H}_{\text{eff}, i} = -\frac{\partial E}{\partial \mathbf{M}_i},$$ (S3)

in terms of the energy $E = \langle H \rangle$, the equations of motion for $\mathbf{\mu}_i$ are obtained by expanding

$$\frac{\partial \mathbf{M}_i}{\partial t} = -g \left( \mathbf{M}_i \times \mathbf{H}_{\text{eff}, i} \right),$$ (S4)

to first order in $\mathbf{\mu}_i$:

$$\mathbf{\mu}_i = -g \left\{ M_0 \times \left[ \mathbf{h}_i + \mathbf{J} \cdot (\mathbf{\mu}_{i+1} + \mathbf{\mu}_{i-1}) \right] - \kappa \mathbf{\mu}_i \right\},$$ (S5)

which assumes that $n_i = \pm 1$ for each site in the unit cell, i.e., a collinear spin state.

Alternatively, we may directly expand $H$ in powers of $\mathbf{\mu}_i$ to obtain $H = E + H_2 + \ldots$ where

$$H_2 = -\sum_{i=1}^{M} J_{\alpha \beta} \mu_{\alpha i} \mu_{\beta i+1, \beta} - \sum_{i=1}^{M} \mathbf{\mu}_i \cdot \mathbf{h}_i - \frac{\kappa}{2} \sum_{i=1}^{M} \mu_{i}^2 - \frac{1}{8\pi} \sum_{i=1}^{M} h_i^2,$$  
(S6)

If the Lagrangian is written [3] in terms of $\mathbf{\mu}_i$, $\dot{\mathbf{\mu}}_i$, and $\phi_i$ as

$$L = \frac{1}{2gM_0} \sum_{i=1}^{M} (\dot{\mathbf{\mu}}_i \times \mathbf{n}_i) \cdot \mathbf{\mu}_i - H_2,$$  
(S7)

then the Hamiltonian equations of motion for $\mathbf{\mu}_i$ given above can also be obtained from the Euler-Lagrange equations

$$\frac{d}{dt} \frac{\partial L}{\partial \dot{\mathbf{\mu}}_i} = \frac{\partial L}{\partial \mathbf{\mu}_i},$$ (S8)

Based on the Lagrangian $L$, the energy-momentum tensor [3] $T_{\alpha \beta}$ is given by

$$T_{\alpha \beta} = L \delta_{\alpha \beta} - \sum_i \frac{\partial \mathbf{\mu}_i}{\partial x_\alpha} \cdot \frac{\partial L}{\partial (\mathbf{\mu}_i / \partial x_\beta)} - \sum_i \frac{\partial \phi_i}{\partial x_\alpha} \frac{\partial L}{\partial (\phi_i / \partial x_\beta)},$$ (S9)

where $x_\alpha = x, y, z$, or $t$ for $\alpha = 1, 2, 3$, or 4, respectively. It follows that the momentum density $(\alpha \neq 4, \beta = 4)$ is

$$p_\alpha = T_{\alpha 4} = -\sum_i \frac{\partial \mathbf{\mu}_i}{\partial x_\alpha} \cdot \frac{\partial L}{\partial \dot{\mathbf{\mu}}_i} = -\frac{1}{2gM_0} \sum_i (\mathbf{n}_i \times \mathbf{\mu}_i) \cdot \frac{\partial \mathbf{\mu}_i}{\partial x_\alpha}.$$ (S10)

Writing $p_\alpha = \sum_i p_{i\alpha}$ gives the expression for $p_{i\alpha}$ stated in Eq. (1) of the main paper. The momentum $p_\alpha$ then satisfies the continuity relation

$$\frac{dp_\alpha}{dt} + \sum_{\beta=1}^{3} \frac{\partial T_{\alpha \beta}}{\partial x_\beta} = 0$$ (S11)

for $\alpha = 1, 2, 3$.

Transforming to the local reference frame of the spin (in the same spirit as in spin-wave theory [4]), we use the local spin variables $\mathbf{\Pi}_i$ given by

$$\mathbf{\Pi}_{ix} = n_{iz} \mathbf{\mu}_{ix},$$ (S12)
$$\mathbf{\Pi}_{iy} = \mathbf{\mu}_{iy},$$ (S13)
$$\mathbf{\Pi}_{iz} = n_{iz} \mathbf{\mu}_{iz},$$ (S14)

with $\mathbf{\Pi}_i^\pm = \mathbf{\Pi}_{ix} \pm i \mathbf{\Pi}_{iy}$ to find the total OAM

$$\mathcal{L}_z = \sum_{i=1}^{N} (\mathbf{r}_i \times \mathbf{p}_i)_z$$

$$= \frac{1}{4gM_0} \sum_{i=1}^{N} \left\{ \mathbf{\Pi}_i^+ \cdot \mathbf{i}_z \mathbf{\Pi}_i^- - \mathbf{\Pi}_i^- \cdot \mathbf{i}_z \mathbf{\Pi}_i^+ \right\},$$ (S15)
where

$$\hat{L}_{z_1} = -i \left( x_i \frac{\partial}{\partial y_j} - y_i \frac{\partial}{\partial x_j} \right) \tag{S16}$$

is the classical OAM operator in real space.

### SYMMETRY RELATIONS AND THE FM ZIG-ZAG LATTICE

In order to clarify the symmetry relations for $X^{-1}(k)$, we review some details of the OAM solution for the FM zig-zag lattice (case i). Based on the $L(k)$ matrix in Eq. (6) of the main paper, we find that the eigenvectors of $L(k) \cdot N$ are

$$\begin{align*}
X(k)_{ij}^* &= c_i^* (-|\Psi_k|, \Psi_k, 0, 0), \\
X(k)_{ij} &= c_i^2 ([\Psi_k, |\Psi_k|, 0, 0]), \\
X(k)_{ij} &= c_3 (0, 0, -|\Psi_k|, \Psi_k), \\
X(k)_{ij} &= c_4 (0, 0, |\Psi_k|, \Psi_k),
\end{align*} \tag{S17}$$

where

$$\Psi_k = \frac{J_1(e^{-ik_x a} + e^{-ik_y a}) + J_2(e^{ik_x a} + e^{ik_y a})}{2(J_1 + J_2)}. \tag{S21}$$

Hence,

$$X(k) = \begin{pmatrix} -c_1 |\Psi_k| & c_1 \Psi_k^* & 0 & 0 \\
-\Psi_k c_2 & c_2 \Psi_k^* & 0 & 0 \\
0 & 0 & -c_3 \Psi_k & c_3 \Psi_k^* \\
0 & 0 & c_4 \Psi_k & c_4 \Psi_k^* \end{pmatrix} \tag{S22}$$

and

$$X^{-1}(k) = \frac{1}{2\Psi_k^*} \begin{pmatrix} \Psi_k^* c_1 & \Psi_k c_2 & 0 & 0 \\
1/c_1 & 1/c_2 & 0 & 0 \\
0 & 0 & -\Psi_k^* c_3 & \Psi_k^* c_4 \\
0 & 0 & 1/c_3 & 1/c_4 \end{pmatrix}, \tag{S23}$$

where $\Psi_k = |\Psi_k|/|\Psi_k|$. With $M = 2$, the symmetry relations [S4] $X^{-1}(k)_{r+n} = X^{-1}(-k)_{r+M,n+M}$ require that $c_3 = c_1$ and $c_4 = c_2$. In addition,

$$X(k) \cdot N \cdot X^{-1}(k) = N \tag{S24}$$

requires that $|c_1|^2 = |c_2|^2 = 1/2 |\Psi_k|^2$, which produces Eq. (8) in the main paper.

Notice that Eq. (S24) can be rewritten as

$$X^{-1}(k) \cdot N \cdot X^{-1}(k) = N \tag{S25}$$

which leads to

$$\sum_{r=1}^{M} \left| X^{-1}(k)_{r,n} \right|^2 - \left| X^{-1}(k)_{r+M,n} \right|^2 = 1. \tag{S26}$$

This expression allows us to rewrite the general result for the OAM of mode $n$ as

$$L_{zn}(k) = \frac{\hbar}{2} \sum_{r=1}^{M} \left\{ X^{-1}(k)_{r,n} \hat{L}_{z_k} X^{-1}(k)_{r,n}^* \right\}$$

$$-X^{-1}(k)_{r+M,n} \hat{L}_{z_k} X^{-1}(k)_{r+M,n}^* \right\}$$

$$= -\frac{i\hbar}{2} \sum_{r=1}^{M} \left\{ \text{Re} X^{-1}(k)_{r,n} \hat{L}_{z_k} \text{Im} X^{-1}(k)_{r,n} \right\}$$

$$-\text{Im} X^{-1}(k)_{r,n} \hat{L}_{z_k} \text{Re} X^{-1}(k)_{r,n}$$

$$-\text{Re} X^{-1}(k)_{r+M,n} \hat{L}_{z_k} \text{Im} X^{-1}(k)_{r+M,n}$$

$$+ \text{Im} X^{-1}(k)_{r+M,n} \hat{L}_{z_k} \text{Re} X^{-1}(k)_{r+M,n} \right\}. \tag{S27}$$

So $L_{zn}(k)$ vanishes if both matrix elements $X^{-1}(k)_{r,n}$ and $X^{-1}(k)_{r,M,n}$ are real.

Returning to the FM zig-zag model (case i), $\Psi_k$ is complex except when $J_1 = J_2$ and $\Psi_k = (\cos(k_x a) + \cos(k_y a))/2$. Since the matrix elements $X^{-1}(k)_{r,n}$ and $X^{-1}(k)_{r+M,n}$ are then also real, magnons of the square-lattice FM carry no OAM. Similar conclusions follow for magnons of the square-lattice AF.

### ANALYTIC RESULTS FOR THE SPIN-SPIN CORRELATION FUNCTION OF THE AF HONEYCOMB LATTICE

For the AF honeycomb lattice (case iv), it is straightforward to evaluate the spin-spin correlation function using the method in Ref. [S4]. As expected, only the transverse $xx$ and $yy$ matrix elements contribute to $S_{ab}(k, \omega)$ evaluated at the degenerate magnon frequency:

$$S_{ab}(k, \omega) = \delta_{ab} \sum_n S^{(n)}_{ab}(k) \delta(\omega - \omega_n(k)) \tag{S28}$$

where $S^{(n)}_{zz}(k)$ is 0 and

$$S^{(1)}_{xx}(k) = S^{(1)}_{yy}(k) = \frac{S}{4} \left\{ |X^{-1}(k)_{11}|^2 + |X^{-1}(k)_{41}|^2 \right\}$$

$$S^{(2)}_{xx}(k) = S^{(2)}_{yy}(k) = \frac{S}{4} \left\{ |X^{-1}(k)_{22}|^2 + |X^{-1}(k)_{32}|^2 \right\}$$

with $f_k = \sqrt{(1 + \kappa)^2 - |T_k|^2}$. So the spin-spin correlation functions for the major and minor bands are the same.
FIG. S1. Berry curvature of the lower magnon band for (a) a FM zig-zag lattice (case i) with \( J_2/J_1 = 1.5 \), and (b) a FM honeycomb lattice (case iii) with \( d = -2D/3J = 0.0067 \). In panel (a), broken lines and dash-dot lines, respectively, indicate \( k_x - k_y = \pm \pi/a \), where the magnon gap is opened by \( J_2/J_1 \neq 1 \), and \( k_x + k_y = \pm \pi/a \), where the magnon gap is always closed. The Berry curvature diverges along the \( k_x + k_y = \pm \pi/a \) lines.

**COMPARISON WITH BERRY CURVATURE**

The Berry curvature of a multiband system in momentum space \([S5]\) is given by

\[
\Omega_{nk} = \frac{i}{2\pi} \sum_{m \neq n} \left\{ \frac{\langle n | \hat{v}_{x k} | m \rangle \langle m | \hat{v}_{y k} | n \rangle}{(\varepsilon_{nk} - \varepsilon_{nk})^2} \right\} \left\{ \frac{\langle n | \hat{v}_{y k} | m \rangle \langle m | \hat{v}_{x k} | n \rangle}{(\varepsilon_{nk} - \varepsilon_{nk})^2} \right\},
\]

(S31)

where, \( n \) and \( m \) are band indices, \( \varepsilon_{nk} \) is the dispersion of band \( n \), and \( \hat{v}_{x k} \) is the velocity operator. With the Hamiltonian written as \( H = \sum_k \hat{H}_k \), \( \hat{v}_{x k} = \partial \hat{H}_k / \partial k_y \).

In order to highlight the difference between the OAM and the Berry curvature, we consider the FM zig-zag and honeycomb lattice models. For the FM zig-zag lattice model (case i) with \( J_2/J_1 = 1.5 \), the Berry curvature in Fig. S1(a) diverges where the gap closes between two bands at \( k_x - k_y = \pm \pi/a \). To suppress this divergence, a small quantity \( \sim 10^{-4} \) is introduced in the denominators of the gauge-invariant form of \( \Omega_{nk} \) above. Except for this divergence, the Berry curvature is quite flat, with some intensity modulations where the magnon gap is opened by \( J_2/J_1 \neq 1 \) at \( k_x + k_y = \pm \pi/a \). The Berry curvature changes sign at \( k_x + k_y = \pm \pi/a \). As shown in the main text, the magnon OAM for the FM zig-zag lattice with \( J_2/J_1 \neq 1 \) has peak intensity at \( k_x + k_y = \pm \pi/a \) and changes sign across this line at \( k_x = k_y \).

For the FM honeycomb lattice (case iii) with \( d = -2D/3J = 0.0067 \), the Berry curvature in Fig. S1(b) has sixfold symmetry and peaks at the K points. On the other hand, the magnon OAM has threefold symmetry, that is the K and K’ points are different. Thus, while both the magnon OAM and Berry curvature exhibit some topological nature originating from the Berry connection, they capture different aspects of the system topology.

**PERIODIC FUNCTIONS \( \overline{k}_x \) AND \( \overline{k}_y \)**

On a discrete lattice, the continuous derivative \( \partial / \partial x_\alpha \) should be replaced by a finite difference. Fig. S2 sketches the distinct lattice translation vectors \( \mathbf{R}_l \) that couple site 1 to other sites of type 1 for for the zig-zag and honeycomb lattices. The finite difference of a discrete function \( f(x) \) produced by translation vector \( \mathbf{R}_l \) is then

\[
\delta_l f(x) = \frac{1}{2|R_l|} \left\{ f(x + R_l) - f(x - R_l) \right\}. \tag{S32}
\]

The continuous derivative \( \partial f(x) / \partial x_\alpha \) along the \( \alpha \) direction is converted into a finite difference by the summation

\[
\Delta_\alpha f(x) = \sum_l \frac{R_{l\alpha}}{|R_l|} \delta_l f(x) \tag{S33}
\]

\[
\Delta_\alpha f(x) = \sum_l \frac{R_{l\alpha}}{|R_l|^2} \left\{ f(x + R_l) - f(x - R_l) \right\},
\]

where \( R_{l\alpha} \) is the projection of the lattice translation vector \( \mathbf{R}_l \) along the \( \alpha \) axis. Note that the finite difference \( \Delta_\alpha f(x) \) approaches the continuous derivative \( \partial f(x) / \partial x_\alpha \) when the lattice translation vectors are orthogonal and their size vanishes.

The finite difference of the factor \( \exp(i\mathbf{k} \cdot \mathbf{x}) \) that enters the Fourier transform of a magnon annihilation operator is given by

\[
\Delta_\alpha \exp(i\mathbf{k} \cdot \mathbf{x}) = i \exp(i\mathbf{k} \cdot \mathbf{x}) \sum_l \frac{R_{l\alpha}}{|R_l|^2} \sin(k \cdot R_l)
\]

\[
= i \overline{k}_\alpha \exp(i\mathbf{k} \cdot \mathbf{x}), \tag{S34}
\]

where

\[
\overline{k}_\alpha = \sum_l \frac{R_{l\alpha}}{|R_l|^2} \sin(k \cdot R_l).
\]

\[
\tag{S35}
\]
FIG. S3. A comparison between the OAM for the FM zig-zag lattice (case i) with different values of $J_2/J_1$ using periodic (top) and non-periodic (bottom) expressions for $k_x$ and $k_y$. In the lower-left panel, broken lines and dash-dot lines, respectively, indicate $k_x - k_y = \pm \pi/a$, where the magnon gap is open by $J_2/J_1 \neq 1$, and $k_x + k_y = \pm \pi/a$, where the magnon gap is always closed.

A periodic expression for $L_{zn}(k)$ is obtained by using the revised OAM operator

$$\hat{l}_{zk} = -i \left( \vec{k}_x \frac{\partial}{\partial k_y} - \vec{k}_y \frac{\partial}{\partial k_x} \right).$$  \hspace{1cm} (S36)

In the limit $N \to \infty$, the momentum-space derivatives $\partial/\partial k_x$ and $\partial/\partial k_y$ do not need to be replaced by their finite differences.

Figure S3 plots the OAM of the FM zig-zag model (case i) for three values of $J_2/J_1$. Summing over the two translation vectors in Fig. S2(a), we obtain

$$\vec{k}_x a = \sin(k_x a) \cos(k_y a),$$
$$\vec{k}_y a = \sin(k_y a) \cos(k_x a).$$  \hspace{1cm} (S37)

To construct the top three panels, the non-periodic $k_x$ and $k_y$ have been replaced by $\vec{k}_x$ and $\vec{k}_y$ in the OAM operator $\hat{l}_{zk}$. When non-periodic functions $k_x$ and $k_y$ are retained in the OAM operator, the OAM plotted in the bottom three panels of Fig. S3 is not bounded and has a much larger magnitude than in the top three panels. Hence, the periodic functions $\vec{k}_x$ and $\vec{k}_y$ impose a bound on the OAM.

For the AF zig-zag lattice with four distinct lattice sites, summing over two translation vectors in Fig. S2(b) gives the periodic wavevectors

$$\vec{k}_x a = \frac{1}{2} \sin(k_x a - k_y a) + \frac{1}{4} \sin(2k_x a + 2k_y a),$$
$$\vec{k}_y a = -\frac{1}{2} \sin(k_x a - k_y a) + \frac{1}{4} \sin(2k_x a + 2k_y a).$$  \hspace{1cm} (S38)

For the FM or AF honeycomb lattice, summing over the three translation vectors in Fig. S2(b) gives

$$\vec{k}_x a = \sin(3k_x a/2) \cos(\sqrt{3}k_y a/2),$$
$$\vec{k}_y a = \frac{1}{\sqrt{3}} \left\{ \sin(\sqrt{3}k_y a/2) \cos(3k_x a/2) + \sin(\sqrt{3}k_y a) \right\}.$$  \hspace{1cm} (S39)

In the limit of small $k_x$ and $k_y$, $\vec{k}_x \to k_x$ and $\vec{k}_y \to k_y$ for the zig-zag lattices while $\vec{k}_x \to 3k_x/2$ and $\vec{k}_y \to 3k_y/2$ for the honeycomb lattices.

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