

Chiral magnon in ferromagnetic chiral crystals

Dapeng Yao¹ and Takehito Yokoyama¹

¹*Department of Physics, Tokyo Institute of Technology,
2-12-1 Ookayama, Meguro-ku, Tokyo 152-8551, Japan*

We theoretically propose chiral magnon in ferromagnetic chiral crystals. We show that the crystal chirality is imprinted in orbital angular momentum of magnons which exhibits the opposite signs for opposite chiralities of the crystal. We also show that a finite magnon orbital angular momentum can be induced by a temperature gradient which is a magnonic analogue of the Edelstein effect.

Chirality is a fundamental property of an object not identical to its mirror image by breaking of reflection or inversion symmetries. Chirality of a lattice structure, which has either a right-handed or left-handed helix, not only leads to the chirality-induced spin selectivity effect [1–4] on the spin degrees of freedom, but also induces an orbital magnetization by an electric current [5, 6]. Crystals with chirality exist in nature such as tellurium or selenium. Due to the low crystallographic symmetries, electricity and magnetism can be coupled in these chiral materials, leading to novel orbital effects. For instance, a current-induced orbital magnetization has been measured in nonmagnetic elemental tellurium [7, 8], and an hedgehog orbital texture can be found in *p*-type tellurium [9].

Chiral phonons characterized by circular motions of ions carry phonon angular momentum, and they possess chirality due to the low-symmetry crystal structure [10–13]. Such circular rotations also emerge in a three-dimensional (3D) chiral crystal and propagate along the screw chain [14]. Moreover, various unconventional physics related to phonon angular momentum have been investigated, such as couplings between chiral phonons, electrons, and magnons [15–20], the phonon Edelstein effect [21], the phonon rotoelectric effect [22], the chiral phonon diode effect [23], and chiral phonon-induced spin current [24–26].

Similar to orbital effects of electrons or phonons, magnons as magnetically collective excitations show an orbital angular momentum, which results in a macroscopic orbital magnetization [27]. In the recent works, the orbital angular momentum (OAM) of magnons has been explicitly formulated in collinear magnets [28–30]. Different from spin magnetization as the projection of spins onto the quantization axis, OAM of magnons derives from the rotation of the perpendicular components of spins in analogy with phonon angular momentum [28, 29]. Recent works on magnon OAM include OAM of the twisted magnonic beams [31], generation of magnon OAM by a skyrmion-textured domain wall in a ferromagnetic nanotube [32], and intrinsic magnon orbital Hall effect [33].

In this Letter, considering the interplay between exchange interactions and a chiral crystal structure, we introduce a spin model with a chiral exchange interaction in a 3D chiral crystal with chirality. Then, we theoretically propose a chiral magnon with an OAM. The crystal

chirality is imprinted in OAM of chiral magnons. In the presence of time-reversal symmetry, the total magnon OAM as a summation over the whole Brillouin zone vanishes in equilibrium because the magnon OAM of each mode is an odd function of the wavevector $\mathbf{k}$ by time-reversal symmetry [28–30]. Nevertheless, we show that a finite magnon OAM can be induced by a temperature gradient, leading to a magnon orbital Edelstein effect (MOEE). This effect is an orbital version of the magnonic analog of the Edelstein effect [34], and analog of the Edelstein effect in electric [5, 6, 35] or phononic [21] systems.

In order to combine the crystal chirality and ferromagnetism, we consider a 3D chiral crystal structure composed of infinitely stacked two-dimensional (2D) honeycomb lattice layers as shown in Fig. 1 [5, 6]. The crystal structures have two distinguishable helices with the left-handed helix in Fig. 1(a) and the right-handed helix in Fig. 1(b), and they can be changed into each other by the mirror reflection M_x with respect to the yz plane. Here, we discuss the case of the right-handed helix as an example. The detailed description of these two crystal structures is included in the Supplemental Materials [36]. For the chiral crystal with the right-handed helix, we label the nearest-neighboring vectors as δ_i ($i = 1, 2, 3$) and the next nearest-neighboring vectors as $\mathbf{R}_i$ ($i = 1, 2, 3$) as shown in Fig. 1(c). Next, we consider the vectors connecting the atoms in the same sublattice between two layers which correspond to chiral spin-spin interactions. In the right-handed helix, the interlayer vectors between A sublattices are $\pm(\mathbf{R}_i + c\hat{z})$, and those between B sublattices are $\pm(-\mathbf{R}_i + c\hat{z})$ as shown in Fig. 1(b).

We now introduce a ferromagnetic Heisenberg spin Hamiltonian with exchange interactions for the chiral crystals, which is given by

$$H = -J_1 \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j - J_2 \sum_{[ij]} \mathbf{S}_i \cdot \mathbf{S}_j, \quad (1)$$

where the first term denotes the nearest-neighboring exchange interaction within the honeycomb layers with the exchange interaction $J_1 > 0$, and the second term represents the chiral exchange interaction between the two layers with exchange parameter $J_2 > 0$ as shown in Figs. 1(a) and 1(b). Then we introduce the ladder operators $S_i^\pm = S_i^x \pm iS_i^y$, and by means of the Holstein-Primakoff transformation [37]: $S_i^z = S - a_i^\dagger a_i$, $S_i^+ \approx \sqrt{2S}a_i$, and $S_i^- \approx \sqrt{2S}a_i^\dagger$ with the magnitude of

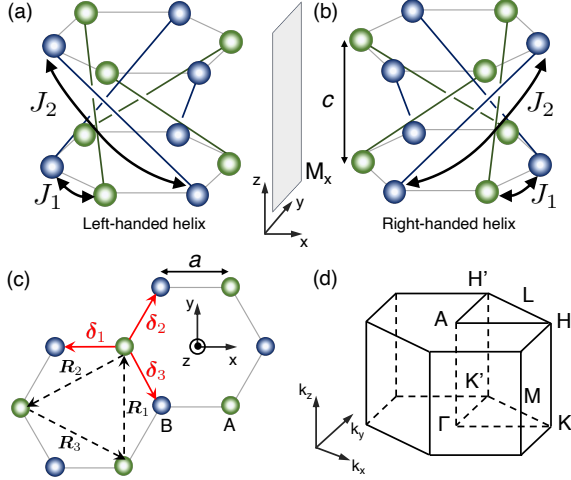


FIG. 1. (Color online) Chiral crystals composed of a stacked honeycomb lattice with (a) left-handed helix and (b) right-handed helix. They can be reflected to each other by the mirror operation M_x with respect to yz plane. Lattice constant along the z direction is denoted by c . The parameter J_1 is the nearest-neighbor exchange interaction within the 2D layers, and J_2 is the chiral exchange interaction between the 2D layers. (c) 2D honeycomb layer. Solid arrows represent the vectors connecting the nearest-neighboring sites: $\delta_1 = a(-1, 0)$, $\delta_2 = a(1/2, \sqrt{3}/2)$, and $\delta_3 = a(1/2, -\sqrt{3}/2)$ with a being the bond length. Dashed arrows denote the vectors connecting the next nearest-neighboring sites: $R_1 = a(0, \sqrt{3})$, $R_2 = a(-3/2, -\sqrt{3}/2)$, and $R_3 = a(3/2, -\sqrt{3}/2)$. (d) First Brillouin zone of the chiral crystal.

spin S , the spin Hamiltonian in Eq. (1) can be expressed in a bosonic formulation as

$$H = -J_1 S \sum_{\langle ij \rangle} (a_i^\dagger a_j + \text{h.c.}) - J_2 S \sum_{[ij]} (a_i^\dagger a_j + \text{h.c.}) + 3(J_1 + 2J_2) S \sum_i a_i^\dagger a_i, \quad (2)$$

with $a_i^\dagger (a_i)$ denoting the creation (annihilation) operator of magnons. Here, we show the spin Hamiltonian for the right-handed helix as an example, and that for the left-handed helix can be obtained by the mirror reflection M_x . The details of the derivation are given in the Supplemental Materials [36]. For the right-handed helix, the Hamiltonian can be rewritten in terms of a quadratic Bogoliubov-de Gennes (BdG) formulation as $H = \sum_{\mathbf{k}} \mathbf{v}_{\mathbf{k}}^\dagger \mathcal{H}(\mathbf{k}) \mathbf{v}_{\mathbf{k}}$, where the vector operator is $\mathbf{v}_{\mathbf{k}} = (a_{\mathbf{k},A}, a_{\mathbf{k},B}, a_{-\mathbf{k},A}^\dagger, a_{-\mathbf{k},B}^\dagger)^T$, and the Bloch Hamiltonian becomes

$$\mathcal{H}(\mathbf{k}) = \frac{3}{2}(J_1 + 2J_2)S \begin{pmatrix} 1 - F_{\mathbf{k}} & -\Gamma_{\mathbf{k}}^* & 0 & 0 \\ -\Gamma_{\mathbf{k}} & 1 - G_{\mathbf{k}} & 0 & 0 \\ 0 & 0 & 1 - F_{\mathbf{k}} & -\Gamma_{\mathbf{k}}^* \\ 0 & 0 & -\Gamma_{\mathbf{k}} & 1 - G_{\mathbf{k}} \end{pmatrix}, \quad (3)$$

with

$$\Gamma_{\mathbf{k}} = \frac{J_1}{3(J_1 + 2J_2)} \left\{ e^{ik_x a} + 2e^{-i\frac{k_x a}{2}} \cos\left(\frac{\sqrt{3}k_y a}{2}\right) \right\}, \quad (4)$$

$$F_{\mathbf{k}} = \frac{2J_2}{3(J_1 + 2J_2)} \left\{ \cos(\sqrt{3}k_y a + k_z c) + 2 \cos\left(\frac{3k_x a}{2}\right) \cos(\sqrt{3}k_y a - k_z c) \right\}, \quad (5)$$

$$G_{\mathbf{k}} = \frac{2J_2}{3(J_1 + 2J_2)} \left\{ \cos(\sqrt{3}k_y a - k_z c) + 2 \cos\left(\frac{3k_x a}{2}\right) \cos(\sqrt{3}k_y a + k_z c) \right\}. \quad (6)$$

The corresponding magnon energies of the lower and upper modes are given by

$$E_{1(2)\mathbf{k}} = 3(J_1 + 2J_2)S \left\{ 1 - \frac{1}{2}C_{\mathbf{k}} \mp \frac{1}{2}\sqrt{B_{\mathbf{k}}^2 + 4|\Gamma_{\mathbf{k}}|^2} \right\}, \quad (7)$$

where $B_{\mathbf{k}} = G_{\mathbf{k}} - F_{\mathbf{k}}$ and $C_{\mathbf{k}} = G_{\mathbf{k}} + F_{\mathbf{k}}$.

Next, we calculate the magnon OAM by following the method proposed in Refs. [28–30]. Here, we first find the inverse of the paraunitary matrix $X^{-1}(\mathbf{k})$ determined by diagonalizing $\mathcal{H}(\mathbf{k}) \cdot N$, where $N = \sigma_z \otimes I_{2 \times 2}$ is a 4×4 matrix with a 2×2 identical matrix $I_{2 \times 2}$ and the Pauli matrix σ_z , and introduce $\mathbf{v}_{\mathbf{k}} = X^{-1}(\mathbf{k})\mathbf{w}_{\mathbf{k}}$ with $\mathbf{w}_{\mathbf{k}}$ denoting the vector operator in terms of the interacting Boson operators [28–30]. Under the normalization condition $X(\mathbf{k}) \cdot N \cdot X^\dagger(\mathbf{k}) = N$, we can obtain

$$X^{-1}(\mathbf{k}) = \frac{1}{\sqrt{2}\Gamma_{\mathbf{k}}^*} \begin{pmatrix} \Gamma_{\mathbf{k}}^* K_{\mathbf{k}}^- & -\Gamma_{\mathbf{k}}^* K_{\mathbf{k}}^+ & 0 & 0 \\ |\Gamma_{\mathbf{k}}| K_{\mathbf{k}}^+ & |\Gamma_{\mathbf{k}}| K_{\mathbf{k}}^- & 0 & 0 \\ 0 & 0 & \Gamma_{\mathbf{k}}^* K_{\mathbf{k}}^- & -\Gamma_{\mathbf{k}}^* K_{\mathbf{k}}^+ \\ 0 & 0 & |\Gamma_{\mathbf{k}}| K_{\mathbf{k}}^+ & |\Gamma_{\mathbf{k}}| K_{\mathbf{k}}^- \end{pmatrix}, \quad (8)$$

where

$$K_{\mathbf{k}}^\pm = \sqrt{1 \pm \frac{B_{\mathbf{k}}}{\sqrt{B_{\mathbf{k}}^2 + 4|\Gamma_{\mathbf{k}}|^2}}}. \quad (9)$$

Details of this derivation are given in Supplemental Material [36].

Here, the magnon OAM can be calculated by using the inverse of the paraunitary matrix $X^{-1}(\mathbf{k})$ derived from the BdG Hamiltonian in Eq. (3) [28–30]. The expectation value of the OAM as a function of wavevector $\mathbf{k}$ in the n -th eigenmode for the chiral crystal with the right-handed

helix is given by

$$\mathcal{L}_{zn}(\mathbf{k}) = \frac{\hbar}{2} \sum_{n=1}^2 \left\{ X^{-1}(\mathbf{k})_{rn} \hat{l}_{z\mathbf{k}} X^{-1}(\mathbf{k})_{rn}^* - X^{-1}(\mathbf{k})_{r+2,n} \hat{l}_{z\mathbf{k}} X^{-1}(\mathbf{k})_{r+2,n}^* \right\}, \quad (10)$$

where $\hat{l}_{z\mathbf{k}} = -i(\bar{k}_x \partial_{k_y} - \bar{k}_y \partial_{k_x})$ represents the operator of magnon OAM with the periodic functions $\bar{k}_x a = \sin(3k_x a/2) \cos(\sqrt{3}k_y a/2)$ and $\sqrt{3}\bar{k}_y a = \sin(\sqrt{3}k_y a/2) \cos(3k_x a/2) + \sin(\sqrt{3}k_y a)$ within the 2D honeycomb layers to guarantee the periodicity of magnon OAM on a discrete lattice [28, 29]. Then, the magnon OAM in Eq. (10) can be further expressed as

$$\mathcal{L}_{z1(2)}(\mathbf{k}) = \frac{\hbar}{4} \left(1 \pm \frac{B_{\mathbf{k}}}{\sqrt{B_{\mathbf{k}}^2 + 4|\Gamma_{\mathbf{k}}|^2}} \right) \frac{\Gamma_{\mathbf{k}}}{|\Gamma_{\mathbf{k}}|} \hat{l}_{z\mathbf{k}} \frac{\Gamma_{\mathbf{k}}^*}{|\Gamma_{\mathbf{k}}|}, \quad (11)$$

for the lower and upper bands by means of Eq. (8). Here, we notice that the first term

$$\mathcal{L}_0(\mathbf{k}) = \frac{\hbar}{4} \frac{\Gamma_{\mathbf{k}}}{|\Gamma_{\mathbf{k}}|} \hat{l}_{z\mathbf{k}} \frac{\Gamma_{\mathbf{k}}^*}{|\Gamma_{\mathbf{k}}|} \quad (12)$$

denotes the contribution from the 2D honeycomb layers, which is a function of wavenumbers k_x and k_y , and has been thoroughly investigated by the previous studies [28–30]. On the other hand, the second term

$$\Delta\mathcal{L}_{z1(2)}(\mathbf{k}) = \pm \frac{\hbar}{4} \frac{B_{\mathbf{k}}}{\sqrt{B_{\mathbf{k}}^2 + 4|\Gamma_{\mathbf{k}}|^2}} \frac{\Gamma_{\mathbf{k}}}{|\Gamma_{\mathbf{k}}|} \hat{l}_{z\mathbf{k}} \frac{\Gamma_{\mathbf{k}}^*}{|\Gamma_{\mathbf{k}}|}, \quad (13)$$

which comes from the chiral exchange interaction is a function of wavenumbers k_x , k_y and k_z , and plays an essential role in chiral magnons. Figure 2 show the distribution of $\Delta\mathcal{L}_{z2}(\mathbf{k})$ in the k_x - k_y momentum space for $k_z = \pm\pi/3$. We see that $\Delta\mathcal{L}_{z2}(\mathbf{k})$ is six fold symmetric around the z -axis and odd in k_z .

The numerical results of the $\mathbf{k}$ -dependent magnon OAM distributions $\mathcal{L}_{zn}(\mathbf{k})$ in Eq. (11) of each magnon band along the high-symmetry points for the left-handed and right-handed helices are shown in Fig. 3(a) and Fig. 3(b), respectively. One can clearly see that the magnon OAM has the opposite signs between the chiral crystals with the opposite helices. Here, for a given wavevector $\mathbf{k}$, the opposite signs of the magnon OAMs mean that the rotations of the spins around the z axis are in the opposite directions for different helices. Therefore, crystal chirality is imprinted in magnon OAM in chiral crystals. In addition to magnon OAM, spin angular momentum plays an important role in ferromagnets. However, here the spin angular momentum has no difference between the two chiral structures because they can be reflected to each other via the mirror operation M_x : $U\mathcal{H}^R(k_x, k_y, k_z)U^{-1} = \mathcal{H}^L(-k_x, k_y, k_z)$, where $\mathcal{H}^R(\mathbf{k})$ and $\mathcal{H}^L(\mathbf{k})$ are the Bloch spin Hamiltonians for the right-handed and left-handed helices, respectively, where the

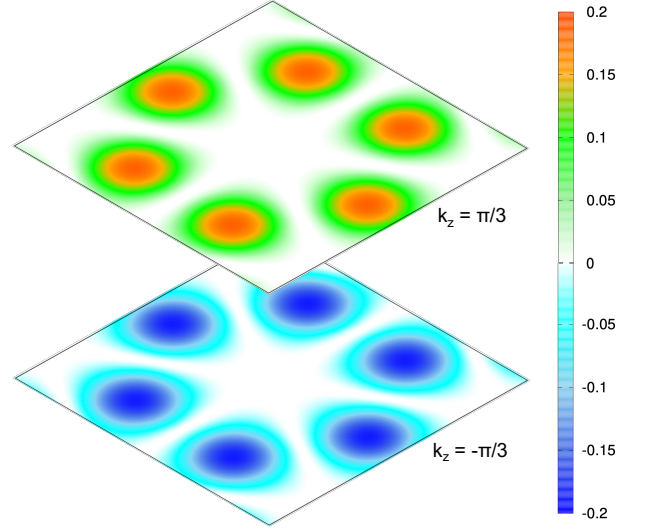


FIG. 2. (Color online) Distribution of $\Delta\mathcal{L}_{z2}(\mathbf{k})$ in Eq. (13) in the k_x - k_y plane. Color represents the magnon OAM in the unit of $\hbar$. The parameters are set to be $J_2 = 0.5J_1$.

unitary matrix is given by $U = I_{2 \times 2} \otimes \sigma_x$ in our model. Thus, the expectation values of the spin along the quantization axis satisfy $\langle S_z^L \rangle = \langle S_z^R \rangle$.

In equilibrium, the total magnon OAM for the z component per volume V is given by

$$L_z^{\text{orb}} = \frac{1}{V} \sum_{\mathbf{k}, n=1,2} \mathcal{L}_{zn}(\mathbf{k}) \{2f_0(E_{n\mathbf{k}}) + 1\}, \quad (14)$$

where $f_0(E_{n\mathbf{k}}) = 1/(e^{E_{n\mathbf{k}}/k_B T} - 1)$ is the Bose-Einstein distribution function [30]. In this case, the total magnon OAM vanishes because time-reversal symmetry requires $X^{-1}(-\mathbf{k}) = X^{-1}(\mathbf{k})^*$, resulting in $\mathcal{L}_{zn}(\mathbf{k}) = -\mathcal{L}_{zn}(-\mathbf{k})$ [28–30]. Nevertheless, a finite magnon OAM can be generated by a temperature gradient because the magnon distribution can become out of equilibrium.

In analogy with the Edelstein effect in phononic systems [21, 23], here we theoretically show that a chiral magnon carrying an OAM can be driven by a temperature gradient in chiral crystals. This phenomenon can be considered as MOEE. In general, a non-equilibrium magnon distribution can be obtained when a temperature gradient $\partial T/\partial x_i$ is applied. By using the Boltzmann equation with the relaxation time approximation, the distribution function of magnons in nonequilibrium is given by

$$f_{n\mathbf{k}} = f_0(E_{n\mathbf{k}}) - \tau v_{n\mathbf{k},i} \frac{\partial f_0}{\partial T} \frac{\partial T}{\partial x_i}, \quad (15)$$

where τ represents the magnon relaxation time and $v_{n\mathbf{k},i} = \partial E_{n\mathbf{k}}/\hbar \partial k_i$ is the group velocity of the n -th mode of magnons. Accordingly, the magnon OAM per unit vol-

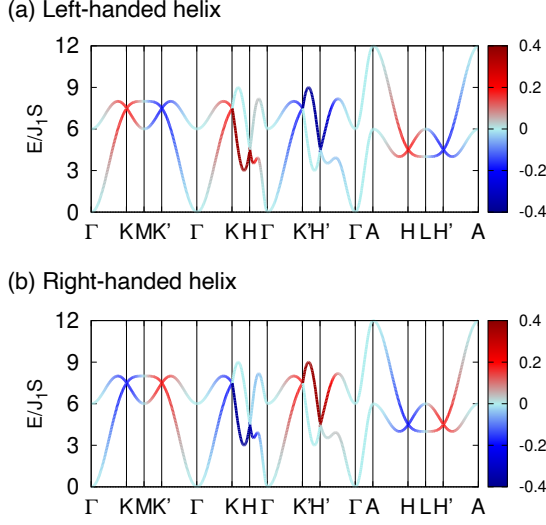


FIG. 3. (Color online) Magnon spectra for the (a) left-handed helix and (b) right-handed helix with the color representing the magnon OAM $\mathcal{L}_{z1(2)}$ in Eq. (11) in the unit of $\hbar$. The parameters are set to be $J_2 = 0.5J_1$.

ume generated by the temperature gradient becomes

$$L_i^{\text{orb}} = -\frac{2\tau}{\hbar V} \sum_{n\mathbf{k}} \mathcal{L}_{in}(\mathbf{k}) \frac{\partial E_{n\mathbf{k}}}{\partial k_j} \frac{\partial f_0(E_{n\mathbf{k}})}{\partial T} \frac{\partial T}{\partial x_j} \equiv \alpha_{ij} \frac{\partial T}{\partial x_j}, \quad (16)$$

where α_{ij} denotes the response tensor, which can be determined by the point-group symmetry. In our model, for the chiral crystals with the D_6 point group, the response tensor is generally represented as [21]

$$\alpha_{ij} = \begin{pmatrix} \alpha_{xx} & 0 & 0 \\ 0 & \alpha_{xx} & 0 \\ 0 & 0 & \alpha_{zz} \end{pmatrix}. \quad (17)$$

Therefore, in the case of the chiral crystals shown in Figs. 1(a) and 1(b), when the temperature gradient is applied along the z direction, we can obtain a finite z component of magnon OAM: $L_z^{\text{orb}} = \alpha_{zz} \partial_z T$, with the temperature gradient $\partial_z T$ and the response coefficient α_{zz} of the MOEE. Here the $\mathbf{k}$ -dependent magnon OAM satisfies $\mathcal{L}_{zn}(k_x, k_y, k_z) = \mathcal{L}_0(k_x, k_y) + \Delta\mathcal{L}_{zn}(k_x, k_y, k_z)$ as given in Eqs. (11), (12), and (13). We notice that the summation in Eq. (16) for the first term $\mathcal{L}_0(k_x, k_y)$ becomes zero due to the time-reversal symmetry. $\mathcal{L}_0(k_x, k_y)$ is independent of k_z , and corresponds to the spin waves only propagating within the 2D honeycomb layer for a given wavevector $\mathbf{k}$. As a result, only $\Delta\mathcal{L}_{zn}(k_x, k_y, k_z)$ contributes to the MOEE coefficient α_{zz} , and α_{zz} eventually becomes

$$\alpha_{zz} = -\frac{2\tau}{\hbar V} \sum_{\mathbf{k}, n=1,2} \Delta\mathcal{L}_{zn}(\mathbf{k}) \frac{\partial E_{n\mathbf{k}}}{\partial k_z} \frac{\partial f_0(E_{n\mathbf{k}})}{\partial T}, \quad (18)$$

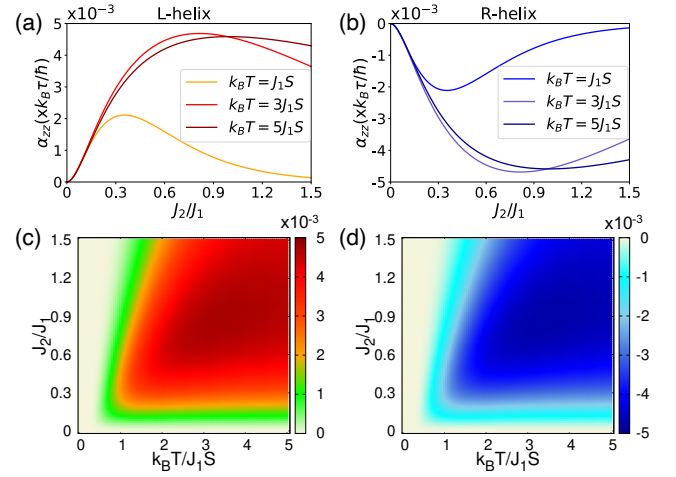


FIG. 4. (Color online) Calculated MOEE coefficient α_{zz} given in Eq. (18). (a) and (b) show α_{zz} versus chiral exchange interaction J_2 with different temperatures for the left-handed and right-handed helices, respectively. (c) and (d) show α_{zz} as a function of $k_B T/J_1 S$ and J_2/J_1 for the left-handed and right-handed helices, respectively.

for the chiral crystals. Figure 4 shows the numerical results of the MOEE coefficient α_{zz} based on Eq. (18). We see that the MOEE coefficient α_{zz} is not monotonic in T and J_2 . The sign of α_{zz} becomes opposite between the crystal structures with the opposite helices because $\Delta\mathcal{L}_{zn}(k_x, k_y, k_z)$ changes the sign when the helix is changed into its mirror image as shown in Fig. 3. Here we notice that the chiral exchange interaction in Eq. (1) contributes to the chirality of magnon OAM.

Let us estimate the size of the magnon OAM generated by a temperature gradient. Here, we set the parameters $J_2 = 0.5J_1$ with $J_1 = 1\text{meV}$, $k_B T = 2.5J_1 S$, and a lattice constant $a = 10^{-9}\text{m}$. The MOEE coefficient is estimated as $\alpha_{zz} \sim 10^{26} \hbar \times [\tau/(1\text{s})] \text{m}^{-2} \text{K}^{-1}$. We use the relaxation time of magnon $\tau = 10^{-10}\text{s}$ and a temperature gradient $\partial_z T = 10\text{K/mm}$ [34]. Then, we can estimate the magnon OAM generated along the z direction with the size of $10^{14} \hbar/\text{cm}^3$, which is of the same order as the magnon Edelstein effect due to the spin angular momentum [34] and experimentally observable.

To summarize, we have theoretically proposed chiral magnon with OAM, which exhibits a chirality in a ferromagnetic chiral crystal due to the lack of inversion and mirror symmetries. A finite magnon orbital angular momentum can be generated by a temperature gradient, leading to MOEE. Therefore, chiral crystals provide a platform for detecting the magnon OAM via the magnon orbital Edelstein effect. We propose a candidate material Cu_2OSeO_3 , which is a magnetic insulator with a chiral structure [38, 39]. Because temperature gradient leads to a redistribution of electrons in metals, this insulating material is suitable for future measurement.

Acknowledgment.— D.Y. was supported by JSPS

-
- [1] K. Ray, S. P. Ananthavel, D. H. Waldeck, and R. Naaman, *Science* **283**, 814 (1999).
- [2] B. Göhler, V. Hamelbeck, T. Z. Markus, M. Kettner, G. F. Hanne, Z. Vager, R. Naaman, and H. Zacharias, *Science* **331**, 894 (2011).
- [3] R. Naaman and D. H. Waldeck, *J. Phys. Chem. Lett.* **3**, 2178 (2012).
- [4] R. Naaman, and D. H. Waldeck, *Annual Review of Physical Chemistry* **66**, 263 (2015).
- [5] T. Yoda, T. Yokoyama, and S. Murakami, *Scientific Reports* **5**, 12024 (2015).
- [6] T. Yoda, T. Yokoyama, and S. Murakami, *Nano Lett.* **18**, 916 (2018).
- [7] T. Furukawa, Y. Shimokawa, K. Kobayashi, and T. Itou, *Nat. Commun.* **8**, 954 (2017).
- [8] T. Furukawa, Y. Watanabe, N. Ogasawara, K. Kobayashi, and T. Itou, *Phys. Rev. Res.* **3**, 023111 (2021).
- [9] G. P. Maruggi, J. Ferreira, E. Baggio-Saitovitch, C. Enderlein, and M. B. Silva Neto, *Phys. Rev. Mater.* **7**, 014204 (2023).
- [10] L. Zhang and Q. Niu, *Phys. Rev. Lett.* **112**, 085503 (2014).
- [11] L. Zhang and Q. Niu, *Phys. Rev. Lett.* **115**, 115502 (2015).
- [12] H. Zhu, J. Yi, M.-Y. Li, J. Xiao, L. Zhang, C.-W. Yang, R.A. Kaindl, L.-J. Li, Y. Wang, and X. Zhang, *Science* **359**, 579 (2018).
- [13] T. Zhang and S. Murakami, *Phys. Rev. Res.* **4**, L012024 (2022).
- [14] K. Ishito, H. Mao, Y. Kousaka, Y. Togawa, S. Iwasaki, T. Zhang, S. Murakami, J.-I. Kishine, and T. Satoh, *Nat. Phys.* **19**, 35 (2023).
- [15] D. M. Juraschek and N. A. Spaldin, *Phys. Rev. Mat.* **3**, 064405 (2019).
- [16] Y. Ren, C. Xiao, D. Saparov, and Q. Niu, *Phys. Rev. Lett.* **127**, 186403 (2021).
- [17] G. Xiong, Z. Yu, and L. Zhang, *Phys. Rev. B* **105**, 024312 (2022).
- [18] M. Hamada and S. Murakami, *Phys. Rev. Res.* **2**, 023275 (2020).
- [19] D. Yao and S. Murakami, *J. Phys. Soc. Jpn* **93**, 034708 (2024).
- [20] D. Yao and S. Murakami, *Phys. Rev. B* **105**, 184412 (2022).
- [21] M. Hamada, E. Minamitani, M. Hirayama, and S. Murakami, *Phys. Rev. Lett.* **121**, 175301 (2018).
- [22] M. Hamada and S. Murakami, *Phys. Rev. B* **101**, 144306 (2020).
- [23] H. Chen, W. Wu, J. Zhu, Z. Yang, W. Gong, W. Gao, Y. Yang, S. A. Yang, and L. Zhang, *Nano Lett.* **22**, 1688 (2022).
- [24] K. Kim, E. Vetter, L. Yan, C. Yang, Z. Wang, R. Sun, Y. Yang, A. H. Comstock, X. Li, J. Zhou, L. Zhang, W. You, D. Sun, and J. Liu, *Nature Materials* **22**, 322 (2023).
- [25] X. Li, J. Zhong, J. Cheng, H. Chen, H. Wang, J. Liu, D. Sun, L. Zhang, and J. Zhou, *Sci. China-Phys. Mech. Astron* **67**, 237511 (2024).
- [26] D. Yao, M. Mamoru, and T. Yokoyama, *Appl. Phys. Lett.* **124**, 162603 (2024).
- [27] R. R. Neumann, A. Mook, J. Henk, and I. Mertig, *Phys. Rev. Lett.* **125**, 117209 (2020).
- [28] R. S. Fishman, J. S. Gardner, and S. Okamoto, *Phys. Rev. Lett.* **129**, 167202 (2022).
- [29] R. S. Fishman, L. Lindsay, and S. Okamoto, *J. Phys.: Condens. Matter* **35**, 167202 (2023).
- [30] R. S. Fishman, *Phys. Rev. B* **107**, 214434 (2023).
- [31] C. Jia, D. Ma, A. F. Schäffer, and J. Berakdar, *Nat. Commun.* **10**, 2077 (2019).
- [32] S. Lee and S. K. Kim, *Front. Phys.* **10**, 858614 (2022).
- [33] G. Go, D. An, H-W. Lee, S. K. Kim, arXiv:2303.11687.
- [34] B. Li, A. Mook, A. Raeliarijaona, and A. A. Kovalev, *Phys. Rev. B* **101**, 024427 (2020).
- [35] V.M. Edelstein, *Solid State Communications* **73**, 233 (1990).
- [36] See Supplemental Material, for detailed discussion and details of calculations.
- [37] T. Holstein and H. Primakoff, *Phys. Rev.* **58**, 1098 (1940).
- [38] G. Meunier and M. Bertaud, *J. Appl. Cryst.* **9**, 364 (1976).
- [39] S. Seki, X. Z. Yu, S. Ishiwata, and Y. Tokura, *Science* **336**, 198 (2012).

Supplemental Material for “Chiral magnon in ferromagnetic chiral crystals”

Dapeng Yao¹ and Takehito Yokoyama¹

¹*Department of Physics, Tokyo Institute of Technology,
2-12-1 Ookayama, Meguro-ku, Tokyo 152-8551, Japan*

Supplemental Material I. DERIVATION OF THE BLOCH SPIN HAMILTONIAN

Here we show the derivation of the spin Hamiltonian Eq. (2) in the main text for the chiral structures with the right-handed and left-handed helices. In fact, the choice of the unit cell is not unique, and here in order to discuss the results clearly, the unit cells in the crystals with the opposite chirality are set to be mirror image of each others as shown in Fig. S1. Here, δ'_i ($i = 1, 2, 3$) represents the vector connecting the nearest-neighbor sites in Fig. S1(a) for the left-handed helix, and δ_i ($i = 1, 2, 3$) represents that in Fig. S1(b) for the right-handed helix. The chiral structures shown in Figs. 1(a)(b) in the main text consist of honeycomb-lattice layers, stacked along the z axis. Vectors for the chiral exchange interactions between two atoms are combinations of the vectors connecting the next-nearest-neighbor sites $\mathbf{R}_i$ ($i = 1, 2, 3$) and the translation of c along the z axis. The interlayer vectors between A sublattices are $\pm(\mathbf{R}_i + c\hat{z})$, and those between B sublattices are $\pm(-\mathbf{R}_i + c\hat{z})$ in the right-handed coordinate system.

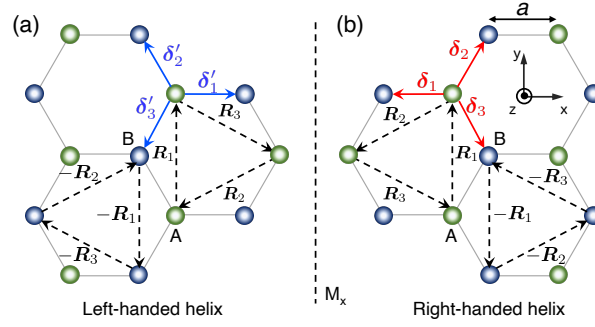


FIG. S1. Schematics of two chiral structures with (a) the left-handed helix and (b) the right-handed helix. The vectors connecting the nearest-neighbor sites in (a) the left-handed helix are $\delta'_1 = a(1, 0)$, $\delta'_2 = a(-1/2, \sqrt{3}/2)$, and $\delta'_3 = a(-1/2, -\sqrt{3}/2)$, and those in (b) the right-handed helix are $\delta_1 = a(-1, 0)$, $\delta_2 = a(1/2, \sqrt{3}/2)$, and $\delta_3 = a(1/2, -\sqrt{3}/2)$.

Then, the first term of the spin Hamiltonian in Eq. (2) for the structure with the left-handed helix can be written as

$$H_1^L = -J_1 S \sum_{\mathbf{r}} \sum_{i=1}^3 a_{B, \mathbf{r} + \delta'_i}^\dagger a_{A, \mathbf{r}} + \text{h.c.} = \sum_{\mathbf{k}} \begin{pmatrix} a_{A, \mathbf{k}}^\dagger & a_{B, \mathbf{k}}^\dagger \end{pmatrix} \begin{pmatrix} 0 & -\gamma_{\mathbf{k}} \\ -\gamma_{\mathbf{k}}^* & 0 \end{pmatrix} \begin{pmatrix} a_{A, \mathbf{k}} \\ a_{B, \mathbf{k}} \end{pmatrix}, \quad (\text{S1})$$

and that for the structure with the right-handed helix can be written as

$$H_1^R = -J_1 S \sum_{\mathbf{r}} \sum_{i=1}^3 a_{B, \mathbf{r} + \delta_i}^\dagger a_{A, \mathbf{r}} + \text{h.c.} = \sum_{\mathbf{k}} \begin{pmatrix} a_{A, \mathbf{k}}^\dagger & a_{B, \mathbf{k}}^\dagger \end{pmatrix} \begin{pmatrix} 0 & -\gamma_{\mathbf{k}}^* \\ -\gamma_{\mathbf{k}} & 0 \end{pmatrix} \begin{pmatrix} a_{A, \mathbf{k}} \\ a_{B, \mathbf{k}} \end{pmatrix}, \quad (\text{S2})$$

where

$$\gamma_{\mathbf{k}} = J_1 S \sum_{i=1}^3 e^{i\mathbf{k} \cdot \delta'_i} = J_1 S \left(\sum_{i=1}^3 e^{i\mathbf{k} \cdot \delta_i} \right)^* = J_1 S \left\{ e^{ik_x a} + e^{-i(k_x a + \sqrt{3}k_y)/2} + e^{-i(k_x a - \sqrt{3}k_y)/2} \right\}, \quad (\text{S3})$$

with $\delta_1 = -\delta'_1$, $\delta_2 = -\delta'_3$, and $\delta_3 = -\delta'_2$. Next, the second term of the spin Hamiltonian in Eq. (2) for the left-handed

helix and that for right-handed helix are the same if we take the vectors in Fig. S1, which is given by

$$\begin{aligned}
H_2 &= -J_2 S \sum_{\mathbf{r}} \sum_{i=1}^3 a_{A,\mathbf{r}+\mathbf{R}_i+c\hat{z}}^\dagger a_{A,\mathbf{r}} + a_{A,\mathbf{r}-\mathbf{R}_i-c\hat{z}}^\dagger a_{A,\mathbf{r}} - J_2 S \sum_{\mathbf{r}} \sum_{i=1}^3 a_{B,\mathbf{r}-\mathbf{R}_i+c\hat{z}}^\dagger a_{B,\mathbf{r}} + a_{B,\mathbf{r}+\mathbf{R}_i-c\hat{z}}^\dagger a_{B,\mathbf{r}} \\
&= -J_2 S \sum_{\mathbf{k}} \begin{pmatrix} a_{A,\mathbf{k}}^\dagger & a_{B,\mathbf{k}}^\dagger \end{pmatrix} \begin{pmatrix} h_{\mathbf{k}}^A & 0 \\ 0 & h_{\mathbf{k}}^B \end{pmatrix} \begin{pmatrix} a_{A,\mathbf{k}} \\ a_{B,\mathbf{k}} \end{pmatrix},
\end{aligned} \tag{S4}$$

with

$$h_{\mathbf{k}}^A = \cos\left(\sqrt{3}k_y a + k_z c\right) + \cos\left(-\frac{3k_x a}{2} - \frac{\sqrt{3}k_y a}{2} + k_z c\right) + \cos\left(\frac{3k_x a}{2} - \frac{\sqrt{3}k_y a}{2} + k_z c\right) \tag{S5}$$

$$h_{\mathbf{k}}^B = \cos\left(\sqrt{3}k_y a - k_z c\right) + \cos\left(-\frac{3k_x a}{2} - \frac{\sqrt{3}k_y a}{2} - k_z c\right) + \cos\left(\frac{3k_x a}{2} - \frac{\sqrt{3}k_y a}{2} - k_z c\right) \tag{S6}$$

The last term of the spin Hamiltonian in Eq. (2) can be written as

$$H_3 = 3(J_1 + 2J_2)S \sum_{\mathbf{k}} \begin{pmatrix} a_{A,\mathbf{k}}^\dagger & a_{B,\mathbf{k}}^\dagger \end{pmatrix} \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} \begin{pmatrix} a_{A,\mathbf{k}} \\ a_{B,\mathbf{k}} \end{pmatrix}. \tag{S7}$$

Therefore, we can write down the total spin Hamiltonian in terms of a quadratic Bogoliubov de Gennes form as $H^\alpha = \sum_{\mathbf{k}} \mathbf{v}_{\mathbf{k}}^\dagger \mathcal{H}^\alpha(\mathbf{k}) \mathbf{v}_{\mathbf{k}}$ with $\alpha = \text{R/L}$, the vector operator $\mathbf{v}_{\mathbf{k}} = \begin{pmatrix} a_{A,\mathbf{k}}^\dagger & a_{B,\mathbf{k}}^\dagger & a_{A,-\mathbf{k}} & a_{B,-\mathbf{k}} \end{pmatrix}^T$, the Bloch Hamiltonian

$$\mathcal{H}^L(\mathbf{k}) = \frac{3}{2}(J_1 + 2J_2)S \begin{pmatrix} 1-F_{\mathbf{k}} & -\Gamma_{\mathbf{k}} & 0 & 0 \\ -\Gamma_{\mathbf{k}}^* & 1-G_{\mathbf{k}} & 0 & 0 \\ 0 & 0 & 1-F_{\mathbf{k}} & -\Gamma_{\mathbf{k}} \\ 0 & 0 & -\Gamma_{\mathbf{k}}^* & 1-G_{\mathbf{k}} \end{pmatrix} \tag{S8}$$

for the left-handed helix and that

$$\mathcal{H}^R(\mathbf{k}) = \frac{3}{2}(J_1 + 2J_2)S \begin{pmatrix} 1-F_{\mathbf{k}} & -\Gamma_{\mathbf{k}}^* & 0 & 0 \\ -\Gamma_{\mathbf{k}} & 1-G_{\mathbf{k}} & 0 & 0 \\ 0 & 0 & 1-F_{\mathbf{k}} & -\Gamma_{\mathbf{k}}^* \\ 0 & 0 & -\Gamma_{\mathbf{k}} & 1-G_{\mathbf{k}} \end{pmatrix}, \tag{S9}$$

for the right-handed helix, where

$$\Gamma_{\mathbf{k}} = \frac{J_1}{3(J_1 + 2J_2)} \left\{ e^{ik_x a} + 2e^{-i\frac{k_x a}{2}} \cos\left(\frac{\sqrt{3}k_y a}{2}\right) \right\}, \tag{S10}$$

$$F_{\mathbf{k}} = \frac{2J_2}{3(J_1 + 2J_2)} \left\{ \cos\left(\sqrt{3}k_y a + k_z c\right) + 2\cos\left(\frac{3k_x a}{2}\right) \cos\left(\sqrt{3}k_y a - k_z c\right) \right\}, \tag{S11}$$

$$G_{\mathbf{k}} = \frac{2J_2}{3(J_1 + 2J_2)} \left\{ \cos\left(\sqrt{3}k_y a - k_z c\right) + 2\cos\left(\frac{3k_x a}{2}\right) \cos\left(\sqrt{3}k_y a + k_z c\right) \right\}. \tag{S12}$$

Supplemental Material II. MAGNON ORBITAL ANGULAR MOMENTUM

In this section, we follow the method proposed by Fishman *et. al.* [1] to calculate the magnon orbital angular momentum (OAM) for the ferromagnetic chiral crystal. Here we show the calculation for the right-handed helix from the Hamiltonian in Eq. (S9) as an example, and that for the left-handed helix from the Hamiltonian in Eq. (S8) can be similarly performed. Based on the Hamiltonian in Eq. (S9), we first find that the eigenvectors of $\mathcal{H}^R(\mathbf{k}) \cdot \mathbf{N}$ are

$$\mathbf{u}_1 = X(\mathbf{k})_{1j}^* = c_1^* (G_{\mathbf{k}} - F_{\mathbf{k}} - \sqrt{4|\Gamma_{\mathbf{k}}|^2 + (G_{\mathbf{k}} - F_{\mathbf{k}})^2} \quad -2\Gamma_{\mathbf{k}} \quad 0 \quad 0), \tag{S13}$$

$$\mathbf{u}_2 = X(\mathbf{k})_{2j}^* = c_2^* (G_{\mathbf{k}} - F_{\mathbf{k}} + \sqrt{4|\Gamma_{\mathbf{k}}|^2 + (G_{\mathbf{k}} - F_{\mathbf{k}})^2} \quad -2\Gamma_{\mathbf{k}} \quad 0 \quad 0), \tag{S14}$$

$$\mathbf{u}_3 = X(\mathbf{k})_{3j}^* = c_3 (0 \quad 0 \quad G_{\mathbf{k}} - F_{\mathbf{k}} - \sqrt{4|\Gamma_{\mathbf{k}}|^2 + (G_{\mathbf{k}} - F_{\mathbf{k}})^2} \quad -2\Gamma_{\mathbf{k}}), \tag{S15}$$

$$\mathbf{u}_4 = X(\mathbf{k})_{4j}^* = c_4 (0 \quad 0 \quad G_{\mathbf{k}} - F_{\mathbf{k}} + \sqrt{4|\Gamma_{\mathbf{k}}|^2 + (G_{\mathbf{k}} - F_{\mathbf{k}})^2} \quad -2\Gamma_{\mathbf{k}}), \tag{S16}$$

where $N = \sigma_z \otimes I_{2 \times 2}$. Hence, the paraunitary matrix of the Hamiltonian in Eq. (S9) can be expressed as

$$X(\mathbf{k}) = \begin{pmatrix} c_1 \left(B_{\mathbf{k}} - \sqrt{B_{\mathbf{k}}^2 + |A_{\mathbf{k}}|^2} \right) & -c_1 A_{\mathbf{k}}^* & 0 & 0 \\ c_2 \left(B_{\mathbf{k}} + \sqrt{B_{\mathbf{k}}^2 + |A_{\mathbf{k}}|^2} \right) & -c_2 A_{\mathbf{k}}^* & 0 & 0 \\ 0 & 0 & c_3^* \left(B_{\mathbf{k}} - \sqrt{B_{\mathbf{k}}^2 + |A_{\mathbf{k}}|^2} \right) & -c_3^* A_{\mathbf{k}}^* \\ 0 & 0 & c_4^* \left(B_{\mathbf{k}} + \sqrt{B_{\mathbf{k}}^2 + |A_{\mathbf{k}}|^2} \right) & -c_4^* A_{\mathbf{k}}^* \end{pmatrix}, \quad (\text{S17})$$

where $A_{\mathbf{k}} = 2\Gamma_{\mathbf{k}}$ and $B_{\mathbf{k}} = G_{\mathbf{k}} - F_{\mathbf{k}}$, and its inverse is

$$X^{-1}(\mathbf{k}) = \frac{1}{2A_{\mathbf{k}}^*} \begin{pmatrix} -\frac{A_{\mathbf{k}}^*}{c_1 \sqrt{B_{\mathbf{k}}^2 + |A_{\mathbf{k}}|^2}} & \frac{A_{\mathbf{k}}^*}{c_2 \sqrt{B_{\mathbf{k}}^2 + |A_{\mathbf{k}}|^2}} & 0 & 0 \\ -\frac{B_{\mathbf{k}} + \sqrt{B_{\mathbf{k}}^2 + |A_{\mathbf{k}}|^2}}{c_1 \sqrt{B_{\mathbf{k}}^2 + |A_{\mathbf{k}}|^2}} & \frac{B_{\mathbf{k}} - \sqrt{B_{\mathbf{k}}^2 + |A_{\mathbf{k}}|^2}}{c_2 \sqrt{B_{\mathbf{k}}^2 + |A_{\mathbf{k}}|^2}} & 0 & 0 \\ 0 & 0 & -\frac{A_{\mathbf{k}}^*}{c_3^* \sqrt{B_{\mathbf{k}}^2 + |A_{\mathbf{k}}|^2}} & \frac{A_{\mathbf{k}}^*}{c_4^* \sqrt{B_{\mathbf{k}}^2 + |A_{\mathbf{k}}|^2}} \\ 0 & 0 & -\frac{B_{\mathbf{k}} + \sqrt{B_{\mathbf{k}}^2 + |A_{\mathbf{k}}|^2}}{c_3^* \sqrt{B_{\mathbf{k}}^2 + |A_{\mathbf{k}}|^2}} & \frac{B_{\mathbf{k}} - \sqrt{B_{\mathbf{k}}^2 + |A_{\mathbf{k}}|^2}}{c_4^* \sqrt{B_{\mathbf{k}}^2 + |A_{\mathbf{k}}|^2}} \end{pmatrix}. \quad (\text{S18})$$

Here the coefficients c_1 , c_2 , c_3 , and c_4 can be obtained by $X(\mathbf{k}) \cdot N \cdot X^\dagger(\mathbf{k}) = N$. Therefore we have

$$\frac{1}{2|c_1|^2} = \frac{1}{2|c_3|^2} = \sqrt{B_{\mathbf{k}}^2 + |A_{\mathbf{k}}|^2} \left(\sqrt{B_{\mathbf{k}}^2 + |A_{\mathbf{k}}|^2} - B_{\mathbf{k}} \right), \quad (\text{S19})$$

$$\frac{1}{2|c_2|^2} = \frac{1}{2|c_4|^2} = \sqrt{B_{\mathbf{k}}^2 + |A_{\mathbf{k}}|^2} \left(\sqrt{B_{\mathbf{k}}^2 + |A_{\mathbf{k}}|^2} + B_{\mathbf{k}} \right), \quad (\text{S20})$$

and finally the inverse of the paraunitary matrix Eq. (S18) becomes

$$X^{-1}(\mathbf{k}) = \frac{1}{\sqrt{2}\Gamma_{\mathbf{k}}^*} \begin{pmatrix} \Gamma_{\mathbf{k}}^* K_{\mathbf{k}}^- & -\Gamma_{\mathbf{k}}^* K_{\mathbf{k}}^+ & 0 & 0 \\ |\Gamma_{\mathbf{k}}| K_{\mathbf{k}}^+ & |\Gamma_{\mathbf{k}}| K_{\mathbf{k}}^- & 0 & 0 \\ 0 & 0 & \Gamma_{\mathbf{k}}^* K_{\mathbf{k}}^- & -\Gamma_{\mathbf{k}}^* K_{\mathbf{k}}^+ \\ 0 & 0 & |\Gamma_{\mathbf{k}}| K_{\mathbf{k}}^+ & |\Gamma_{\mathbf{k}}| K_{\mathbf{k}}^- \end{pmatrix}, \quad (\text{S21})$$

where

$$K_{\mathbf{k}}^\pm = \sqrt{1 \pm \frac{B_{\mathbf{k}}}{\sqrt{B_{\mathbf{k}}^2 + 4|\Gamma_{\mathbf{k}}|^2}}}. \quad (\text{S22})$$

As we mentioned in the main text, the magnon OAM is generally given by Eq. (10) [1–3]. By using the inverse of the paraunitary matrix $X^{-1}(\mathbf{k})$, we can calculate the magnon OAM as

$$\mathcal{L}_{z1}^R(\mathbf{k}) = \frac{\hbar}{2} \left\{ X_{11}^{-1}(\mathbf{k}) \hat{l}_{z\mathbf{k}} X_{11}^{-1}(\mathbf{k})^* + X_{21}^{-1}(\mathbf{k}) \hat{l}_{z\mathbf{k}} X_{21}^{-1}(\mathbf{k})^* \right\} \quad (\text{S23})$$

$$= \frac{\hbar}{2} \left\{ \frac{K_{\mathbf{k}}^-}{\sqrt{2}} \hat{l}_{z\mathbf{k}} \frac{K_{\mathbf{k}}^-}{\sqrt{2}} + \frac{K_{\mathbf{k}}^+}{\sqrt{2}} \frac{|\Gamma_{\mathbf{k}}|}{\Gamma_{\mathbf{k}}^*} \hat{l}_{z\mathbf{k}} \left(\frac{K_{\mathbf{k}}^+}{\sqrt{2}} \frac{|\Gamma_{\mathbf{k}}|}{\Gamma_{\mathbf{k}}} \right) \right\} \quad (\text{S24})$$

$$= \frac{\hbar}{4} \left\{ K_{\mathbf{k}}^+ \hat{l}_{z\mathbf{k}} K_{\mathbf{k}}^+ + K_{\mathbf{k}}^- \hat{l}_{z\mathbf{k}} K_{\mathbf{k}}^- + (K_{\mathbf{k}}^+)^2 \frac{\Gamma_{\mathbf{k}}}{|\Gamma_{\mathbf{k}}|} \hat{l}_{z\mathbf{k}} \frac{\Gamma_{\mathbf{k}}^*}{|\Gamma_{\mathbf{k}}|} \right\} \quad (\text{S25})$$

$$= \frac{\hbar}{4} \left(1 + \frac{B_{\mathbf{k}}}{\sqrt{B_{\mathbf{k}}^2 + 4|\Gamma_{\mathbf{k}}|^2}} \right) \frac{\Gamma_{\mathbf{k}}}{|\Gamma_{\mathbf{k}}|} \hat{l}_{z\mathbf{k}} \frac{\Gamma_{\mathbf{k}}^*}{|\Gamma_{\mathbf{k}}|}, \quad (\text{S26})$$

for the lower band, where $K_{\mathbf{k}}^+ \hat{l}_{z\mathbf{k}} K_{\mathbf{k}}^+ + K_{\mathbf{k}}^- \hat{l}_{z\mathbf{k}} K_{\mathbf{k}}^- = 0$. Similarly, the magnon OAM for the upper band can be calculated as

$$\mathcal{L}_{z2}^R(\mathbf{k}) = \frac{\hbar}{4} (K_{\mathbf{k}}^-)^2 \frac{\Gamma_{\mathbf{k}}}{|\Gamma_{\mathbf{k}}|} \hat{l}_{z\mathbf{k}} \frac{\Gamma_{\mathbf{k}}^*}{|\Gamma_{\mathbf{k}}|} = \frac{\hbar}{4} \left(1 - \frac{B_{\mathbf{k}}}{\sqrt{B_{\mathbf{k}}^2 + 4|\Gamma_{\mathbf{k}}|^2}} \right) \frac{\Gamma_{\mathbf{k}}}{|\Gamma_{\mathbf{k}}|} \hat{l}_{z\mathbf{k}} \frac{\Gamma_{\mathbf{k}}^*}{|\Gamma_{\mathbf{k}}|}. \quad (\text{S27})$$

On the other hand, we can also calculate the magnon OAM for the left-handed helix from the Hamiltonian Eq. (S8):

$$\mathcal{L}_{z1(2)}^L(\mathbf{k}) = \frac{\hbar}{4} (K_{\mathbf{k}}^\pm)^2 \frac{\Gamma_{\mathbf{k}}^*}{|\Gamma_{\mathbf{k}}|} \hat{l}_{z\mathbf{k}} \frac{\Gamma_{\mathbf{k}}}{|\Gamma_{\mathbf{k}}|} = \frac{\hbar}{4} \left(1 \pm \frac{B_{\mathbf{k}}}{\sqrt{B_{\mathbf{k}}^2 + 4|\Gamma_{\mathbf{k}}|^2}} \right) \frac{\Gamma_{\mathbf{k}}^*}{|\Gamma_{\mathbf{k}}|} \hat{l}_{z\mathbf{k}} \frac{\Gamma_{\mathbf{k}}}{|\Gamma_{\mathbf{k}}|}. \quad (\text{S28})$$

Notice that

$$\begin{aligned} \frac{\Gamma_{\mathbf{k}}}{|\Gamma_{\mathbf{k}}|} \hat{l}_{z\mathbf{k}} \frac{\Gamma_{\mathbf{k}}^*}{|\Gamma_{\mathbf{k}}|} &= -\frac{\Gamma_{\mathbf{k}}^*}{|\Gamma_{\mathbf{k}}|} \hat{l}_{z\mathbf{k}} \frac{\Gamma_{\mathbf{k}}}{|\Gamma_{\mathbf{k}}|} \\ &= \frac{-1}{|\gamma_{\mathbf{k}}|^2} \left\{ \sqrt{3} \bar{k}_x \sin K_1 \sin K_2 - \bar{k}_y [(\cos K_1 + 2 \cos K_2)(\cos K_1 - \cos K_2) + \sin^2 K_1] \right\}, \end{aligned} \quad (\text{S29})$$

with $K_1 = 3k_x/2$, $K_2 = \sqrt{3}k_y/2$, and $\gamma_{\mathbf{k}} = \sqrt{1 + 4 \cos K_1 \cos K_2 + 4 \cos^2 K_2}$. Here, we introduce

$$\begin{aligned} \bar{k}_x &= \sin(3k_x a/2) \cos(\sqrt{3}k_y a/2), \\ \bar{k}_y &= \frac{1}{\sqrt{3}a} \left\{ \sin(\sqrt{3}k_y a/2) \cos(3k_x a/2) + \sin(\sqrt{3}k_y a) \right\}, \end{aligned} \quad (\text{S30})$$

corresponding to the 2D honeycomb layers to guarantee the periodicity of magnon OAM on a discrete lattice [1, 2]. Eq. (S29) can have opposite sign if we choose the unit cells to be mirror symmetric as shown in Fig. S1. Thus, we have $\mathcal{L}_{z1(2)}^L(\mathbf{k}) = -\mathcal{L}_{z1(2)}^R(\mathbf{k})$ which reflects the chirality of the magnon OAM in chiral structures.

-
- [1] R. S. Fishman, J. S. Gardner, and S. Okamoto, Phys. Rev. Lett. **129**, 167202 (2022).
 - [2] R. S. Fishman, L. Lindsay, and S. Okamoto, J. Phys.: Condens. Matter **35**, 167202 (2023).
 - [3] R. S. Fishman, Phys. Rev. B **107**, 214434 (2023).