Twisted light-induced spin-spin interaction in a chiral helimagnet

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We theoretically investigate how the orbital angular momentum of light can affect a chiral magnetic order. Here, we consider a metallic chiral helimagnet, which is under stationary radiation of a resonant optical vortex beam. We propose a novel interaction between local spins considering microscopic interactions between an optical vortex and electrons. This vortex-induced interaction modulates the chiral magnetic order in an entirely different way than an external magnetic field does. Our spin modulation technique may pave a route to create a unique topological or chiral structure for future opto-spintronics devices.

Keywords: chiral helimagnet, optical vortex, spintronics

I. INTRODUCTION

Chiral helimagnets are ferromagnetic materials belonging to chiral space groups or non-centrosymmetric space groups. The local spin moments of such magnets are located on the chiral framework of the lattice; thus, its magnetization is twisted. Chiral helimagnetic materials have promising spintronic functionalities for device applications. This is because they have various physical properties owing to nonlinearity, robustness, topology, and tunability [1]. In chiral helimagnet, the chiral spin texture, which means the left- or right-handed helical spin texture along the crystallographic axis, is characterized by the antisymmetric Dzyaloshinskii-Moriya (DM) interaction specified by the DM vector $\vec{D}$ along the helical axis $\vec{D}$ and Heisenberg interaction specified by $J$.

On the other hand, in the aspect of chirality, Allen et al. proposed the twisted light with orbital angular momentum (OAM), so-called optical vortex [4]. The optical vortex carries its intrinsic OAM and has the helical phase structure and toroidal field intensity resulting from the topological singularity along the propagating axis. Recently, many applications of the optical vortex are proposed in broad fields, e.g., stimulated emission depletion microscopy [5], mechanical rotation control of classical micro-particles [6, 7], laser ablation using a chiral beam [8], and shaping some complex shapes of microfibers [9]. Besides, as some attempts to transfer optical OAM to magnetic materials, Fujita et al. proposed an ultrafast generation of skyrmionic defects due to a heat distribution of a vortex beam and a creation of anisotropic spin waves and topological defects in chiral magnets due to a magnetic field of the beam [10, 11].

Theories in magneto-optics since Faraday’s era say that the optical response of magnets stems from transferring the angular momentum of light to that of electrons through the optical transition. Conventionally in this situation, optical angular momentum mostly refers only to spin angular momentum (SAM), i.e., the circular polarization helicity. In analogy with transferring the optical SAM to electrons, the optical OAM can also be transferred via the spin-orbit interaction. Some theoretical and experimental studies show that optical transitions by the vortex are different from those of traditional plane waves [12–14].

We focus on a one-dimensional (1-D) chiral helimagnet, e.g., chromium-intercalated niobium disulfide (CrNb3S6), classified as a metal where itinerant and localized electrons coexist [1]. Various theoretical and experimental works have revealed that the competition between Heisenberg interaction and DM interaction characterizes the chiral magnetic structure [15–19]. The DM interaction in this material acts between the localized spins mediated by the itinerant electrons. This reminds us of the existence of Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction. Although the RKKY interaction is usually invariant for spin rotation, the coexistence of the spin-orbit interaction and the spatially distributed magnetic moments gives rise to the anisotropic effects [20, 21]. In this magnet, the local magnetic moments originating from the OAM of the localized electron in Cr sites polarize the itinerant electrons locally through the spin-orbit interaction. The RKKY picture, where the itinerant electrons are scattered by the local magnetic moment, is sometimes called the generalized RKKY picture [22]. The RKKY interaction with the anisotropic effect causes the DM interaction in this material [1]. The optical vortex and the chiral helimagnetic order have a similar spatial structure from the viewpoint of chirality; a kind of chiral couplings between them can be expected. The RKKY process through

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electron hopping under the optical vortex radiation can affect the interaction between nearest-neighbor local spins nontrivially. We show that this optical vortex-induced spin-spin interaction can modulate the chiral helimagnetic order of CrNb$_3$S$_6$. The interaction creates the possibility of microscopically modulating the axis of rotation of the helical structure, giving the macroscopic helical magnetic structure a new degree of freedom. Such modulation cannot be achieved by the Zeeman effect and magnetic resonance. There are various proposals to control magnetic interactions with electromagnetic fields [23–28]. In addition to these methods, the control of spin-spin interaction by the OAM of light can be a new avenue for the development of magneto-optics or opto-spintronics.

II. MODEL

We model the optical vortex beam by the Laguerre-Gaussian (LG) mode. Here, we assume that the helical magnetic structure and the LG beam are coaxial (see figure 1(a)). The LG beam is represented by the four-vector $A^\mu = (\varphi/c, A^\lambda)$ satisfying the Helmholtz equation. Adopting Lorenz gauge $\partial_\mu A^\mu = 0$, we obtain a traveling wave in the $z$-direction with the frequency $\omega$ and the wave number $k$. It becomes within the paraxial approximation

$$\phi^{(l,s)}(\rho, \eta, z, t) = \frac{e^2}{i \omega} \nabla \cdot A^{(l,s)}(\rho, \eta, z, t),$$

$$A^{(l,s)}(\rho, \eta, z, t) = A_0^l \sqrt{\frac{2}{\pi \ell !}} \frac{1}{w(z)} \left[ \frac{\rho \sqrt{\ell}}{w(z)} \right]^{\ell} \exp \left[ - \frac{\rho^2}{w(z)^2} \right] \exp \left[ i \left( \frac{k \rho^2}{2R(z)} + l \eta - \psi(z) \right) \right] e^{i(kz-\omega t)},$$

where we use the cylindrical coordinate $(\rho, \eta, z) = \left( \sqrt{x^2 + y^2}, \tan^{-1}(y/x), z \right)$, and $A_0^l / |A_0^l|$ represents the polarization of light. Here, $(l, s)$ indicates the OAM and SAM of light, $w(z)$ is the beam radius, $R(z)$ the radius of curvature of the beam and $\psi(z)$ the Gouy phase.

In describing the electronic system in the chiral helimagnet, we treat the itinerant electrons in a multi-orbital tight-binding model, in which they interact with the localized spins via strong Hund’s coupling:

$$H_{ch} = H_0 + H_{hop} + H_{so} = \sum_{i, \mu, \gamma} E_\mu c_{i\mu\gamma}^\dagger c_{i\mu\gamma} - J_H \sum_i \mathbf{S}_i \cdot \mathbf{s}_i + H_{hop} + H_{so},$$

where $E_\mu$ is the energy of $\mu$-electrons. The field operator of the electron is $\psi_{\mu}\mathbf{r} = \sum_{\nu} \phi_{\mu\nu}(\mathbf{r}) c_{\mu\nu}$, where $\phi_{\mu\nu}(\mathbf{r})$ are the Wannier function centered on the site $\mathbf{R}_i$ with $c_{\mu\nu}$ being the corresponding annihilation operator of the itinerant electrons. The indices $\mu$ and $\gamma$ indicate the orbit and spin. The constant $J_H$ denotes the Hund’s coupling energy between the itinerant and localized electron spins. We define the itinerant spin as $s_i = \sum_{\mu, \gamma} c_{i\mu\gamma}^\dagger \sigma_{\mu\gamma} c_{i\mu\gamma}$ with $\sigma$ being Pauli matrix, while the operator $\mathbf{S}_i$ is the localized spin. The hopping and the spin-orbit interaction are

$$H_{hop} = -\Gamma \sum_{i, j, \mu, \nu, \gamma} c_{i\mu\gamma}^\dagger c_{j\nu\gamma},$$

$$H_{so} = \lambda \sum_i \mathbf{L}_i \cdot \mathbf{s}_i,$$

where $\Gamma$ and $\lambda$ are the corresponding coupling constants, and $\mathbf{L}_i$ is the OAM of the ion on the site $\mathbf{R}_i$. Figure 1 shows the schematic of the electronic level structure and the electronic transition by the spin-orbit interaction and optical vortex absorption. We consider only the spin-orbit coupling $L \cdot s_i$ between the ion and itinerant spin. It contributes to excite the itinerant electron from a higher state $e$ to a lower state $a_1$ with spin-flipping (figure 1(b)). The site-dependent $L_i$ corresponds to the local magnetic moment, which reflects the crystal symmetry of the helimagnet. We have neglected the spin-orbit interaction between the local momenta $\mathbf{L}_i \cdot \mathbf{S}_i$. It contributes to the interband transition with spin-flipping whose transition energy is much larger than that by $L_i \cdot s_i$ within the Hund’s rule. In addition, considering the effect of the optical vortex absorption, the $L_i \cdot s_i$ term should be higher-order process in the RKKY scheme.

The 1-D helimagnetic material CrNb$_3$S$_6$ is the transition metal dichalcogenide (NbS$_2$) intercalated with $3d$ magnetic elements (Cr). The localized spin with $S = |\mathbf{S}_i| = 3/2$ is originated from the localized electron in Cr atoms in trivalent state. On the other hand, the itinerant $d$-electrons belong to the NbS$_2$-derived unfilled band. As the Cr atom plays a role of the bridge between the well-separated NbS$_2$ layers, the interlayer electronic conduction occurs via the Cr atom. Thus, the itinerant and localized electrons couple with each other. The local symmetry around Cr is $D_3$ and its $3d$-orbitals split into $e, a_1$ and $e'$ orbitals [29]. Here as the orbital $e$ has $d_{z^2-r^2}$ symmetry and the orbital $a_1$ has $d_{3z^2-r^2}$ symmetry, the interband transition of the d-electron by optical vortices via the dipole interaction is
allowed for \((l, s) = (\pm 1, \pm 1)\) (see figure 1(c)) \([30]\). Note that such an elementary process is forbidden for plane waves with linearly or circularly polarized light due to the transition selection rule, and requires the OAM of light. Thus, we sum over \(a_1\) and \(e\) for the orbitals \(\mu\) and \(\nu\) in the above Hamiltonian.

The ‘vector potential’ in the presence of the spin-orbit interaction is defined by
\[
\chi^{(l, s)}(r, t) = -e A^{(l, s)}(r, t) - \left(\mu_B / 2c^2\right) \sigma \times E^{(l, s)}(r, t) \[31]\]. Here, \(\mu_B\) is the Bohr magneton, and \(E^{(l, s)} = i\omega A^{(l, s)} - \nabla \phi^{(l, s)}\). We introduce the vector potential through the on-site interband excitation, i.e.,
\[
H' = H_{ch} + H^X,
\]
where \(m\) and \(p\) are the mass and momentum of electron. In addition, we have defined the matrix element for some operator \(\mathcal{O}\) as
\[
\langle i\mu\gamma | O | j\nu\gamma' \rangle = \langle \gamma | \int dr \phi_{i\mu}^\dagger(r) O(r) \phi_{j\nu}(r) | \gamma' \rangle.
\]
The form of equation (6) itself does not assume a specific electric field. For instance, when \(E^{(l, s)}(r, t)\) is constant in time and space, it reduces to the spin-orbit interaction of Bychkov-Rashba type \([32]\). Here we consider the oscillating electric field of the optical vortex. The energy of the electric field is resonant with the interband transition, i.e., \(\hbar \omega = E_g - E_{a_1} + S J_H\) is the gap energy including the Hund’s coupling. Thus, one can consider that the itinerant electrons have two spin-orbit interactions, one with the localized moment \(L_i\) at each site and the other with the optical vortex in this system.

**III. EFFECTIVE 1-D SPIN MODEL**

We derive the effective model considering the perturbations with respect to the electron hopping \(H_{\text{hop}}\), spin-orbit interaction \(H_{\text{SO}}\), and optical vortex absorption \(H^X\). The unperturbed Hamiltonian is \(H_0\), and the itinerant electrons...
in the lower band \( (a_i) \) have their spins parallel to the localized spins in the unperturbed ground state. Thus, we impose the constraint not to break the Hund rule throughout the perturbation calculation \cite{33}. Here we consider the time evolution operator in interaction picture

\[
U(t, 0) = T \left\{ \exp \left[ -\frac{i}{\hbar} \int_0^t (H_{\text{hop}} + H_{\text{SO}} + H^x(t')) dt' \right] \right\},
\]

where \( T \) is the time-ordering operator. We expand this operator for each order of the perturbation \( U(\eta, 0) = \sum_{n \geq 0} U^{(n)}(t, 0) \). The Hamiltonian from \( n \)-th perturbation can be written as

\[
H^{(n)}(t) = \sum_{i, j, \gamma, \gamma'} \left\{ i \frac{d}{dt} \langle i, a_1, \gamma | U^{(n)}(t, 0) | j, a_1, \gamma' \rangle \right\} c^\dagger_{i a_1 \gamma} c_{j a_1 \gamma'}.
\]

Throughout the perturbation calculation, we use a so-called secular approximation to neglect components with the fast oscillation \cite{34, 35}. Thus, we can consider only the time-independent Hamiltonian. This approximation is safely justified because the perturbation energies are much smaller than the gap energy \( E_g \). In addition, the period of the neglected oscillation \( \sim 2\pi \hbar / E_g \) is much shorter than the time step of the subsequent simulation. Such an oscillation should be averaged out during the time step.

In the first-order perturbation, only the hopping Hamiltonian \( H_{\text{hop}} \) is relevant. Due to the Hund rule, the localized spins tend to be aligned for the itinerant electrons to have more extended waveforms. Thus, the first-order Hamiltonian makes the localized spins exhibit ferromagnetic tendency, and the exchange coupling \( J \) between the localized spins is proportional to the hopping energy \( \Gamma \) \cite{36}. The details of the perturbation calculations are described in the Supplementary Material. In the second-order perturbation, the products of the spin-orbit interaction and the hopping energy remain. In the third-order perturbation, the triple product of the hopping energy, the spin-orbit interaction, and optical vortex absorption is relevant. By using the method of projection operator, the results of the perturbation calculation can be summarized as follows \cite{36},

\[
H' = H_0 - \Gamma \sum_{i, j, \mu, \nu, \gamma, \gamma'} c^\dagger_{i \mu \gamma} (\gamma | P_i P_j | \gamma') c_{j \nu \gamma'}.
\]

The second term represents the complex one-particle hopping process, which incorporates the effects due to the spin-orbit interaction and the optical vortex absorption. The projection operator at the \( i \)-th site is

\[
P_i = \frac{(S_i + v_i + v'_i) \cdot \sigma + (S + 1)}{2S + 1},
\]

where

\[
v_i = -\frac{i \lambda}{E_g} \langle a_1 | L_i | e \rangle \times S_i,
\]

\[
v'_i = \frac{2i \lambda}{\hbar \delta} \langle a_1 | L_i | e \rangle \times \left( \text{Im} \langle a_1 | B_1^{(l, s)} | e \rangle \times S_i \right).
\]

with \( \delta \) being the phenomenological line width for the optical transition, and \( B_1^{(l, s)} = ( -\mu_B / 2c^2 ) E_1^{(l, s)} \times r_i \). As can be seen from equation (12), the absorption line width \( \delta \) is essential for the third-order perturbation to account for the resonant absorption of the optical vortex. This line width dampens some of the oscillations and allows us to define a time-independent perturbation Hamiltonian at the steady state where \( \exp(-\delta t) \sim 0 \) (see the Supplementary Material). In the absence of the width \( (\delta = 0) \), the perturbation calculation is violated and the time-independent perturbative Hamiltonian is not defined. In addition, we ignore the perturbation terms that contain more than two optical vortex absorption terms. This is because such terms should be small in the parameter regime considered in this study. The three vectors \( S_i, v_i, \) and \( v'_i \) incorporate the effects of the first-, the second-, and the third-order perturbation, respectively. We can obtain the effective Hamiltonian for the localized spins by making the wavefunction of the itinerant electron extend the most in equation (9) \cite{36}.

Mapping the effective Hamiltonian onto 1-D spin model, we obtain the Hamiltonian for the localized ion spin;

\[
\frac{H_{1D}}{S^2} = -J \sum_i \hat{n}_i \cdot \hat{n}_{i+1} + \sum_i D_i \cdot (\hat{n}_i \times \hat{n}_{i+1}) - \sum_i \hat{n}_i \cdot \left( D_i B_1^{(l, s)} + B_1^{(l, s)} D_i \right) \cdot \hat{n}_{i+1},
\]
where \( J \sim \Gamma / S^2 \) is the Heisenberg exchange coupling, and

\[
D_t = -\frac{\lambda J}{E_g} \text{Im} \left( \langle a_1 | L_{i} | e \rangle - \langle a_1 | L_{i+1} | e \rangle \right),
\]

\[
\mathbf{B}^{(l,s)}_t = \frac{2E_g}{\hbar \delta} \text{Im} \left( \langle a_1 | \mathbf{B}^{(l,s)}_1 | e \rangle - \langle a_1 | \mathbf{B}^{(l,s)}_{i+1} | e \rangle \right).
\]

Here \( \mathbf{n}_i \) are the magnitude and the direction of the localized ion spins \((S_i = \mathbf{\hat{n}}_i)\). The DM interaction vector \( \mathbf{D}_t \) originates in the vector \( \mathbf{v}_i \). The RKKY interaction usually depends on the Fermi wavelength. In the present calculation, instead of describing the mediation of conduction electrons by scattering of wave functions, we use a hopping Hamiltonian. In addition, the energies of the initial and intermediate states are described by the band energy without considering the wavenumber dependence. Thus, the Fermi wavelength does not appear in the DM interaction vector. One can consider the differences in local OAMs between adjacent sites are the same everywhere, i.e., \( \mathbf{D}_t = \mathbf{D} \). When the optical vortex-induced term is absent, equation (13) is known to well explain various magnetic features of CrNb_{2}S_{2} with Cr sites twisting [1,2]. In other words, we effectively take into account the helical structure of the Cr's spins through equation (13).

The last term on the right-hand side of equation (13) is the optical vortex-induced spin-spin interaction and originates in the vector \( \mathbf{v}_i \). Note that the vector \( \mathbf{B}^{(l,s)}_i \) does not refer to the magnetic field of light. It is proportional to the LG beam intensity and is the dimensionless vector that determines the coupling between the LG beam and the spins. We assume that the DM vector points in the positive direction of \( \mathbf{L} = \mathbf{D} \hat{e}^z, \mathbf{D} > 0 \) with \( \hat{e}^z \) being the \( z \)-directed unit vector. As for the optical vortex, we assume the LG beam with \((l, s) = (\pm 1, \pm 1)\) and focus on the light-induced effect near the focal spot. We neglect the \( z \)-dependence of \( w(z), R(z), \) and \( \psi(z) \) in the LG beam. In this case, the vector \( \mathbf{B}^{(l,s)}_t = B_0 \sin (ka/2) \mathbf{b}^{(l,s)}_i \) oscillates with \( z \) in the \( x-y \) plane since \( \mathbf{b}^{(\pm 1, \pm 1)} = -\cos [k (z_i + a/2)] \hat{e}^x \pm \sin [k (z_i + a/2)] \hat{e}^y \) (see figure 2). Here, \( a = z_{i+1} - z_i \) is the lattice constant. We rewrite the last term as follows;

\[
-2DB_0 \sin \left( \frac{ka}{2} \right) \sum_i \mathbf{n}_i \cdot \left( \hat{e}^+_i \hat{e}^-_i - \hat{e}^-_i \hat{e}^+_i \right) \cdot \mathbf{n}_{i+1},
\]

where \( \hat{e}^\pm_i = (\mathbf{b}^{(l,s)}_i \pm \hat{e}^z) / \sqrt{2} \). Since the Heisenberg coupling is dominant in this system, this term tends to align the \( \hat{e}^+ \)-components of the adjacent local spins. On the contrary, for the chiral magnet with \( \mathbf{D} < 0 \), the term tends to align the \( \hat{e}^- \)-components. In this sense, one can interpret this vortex-induced interaction as a kind of chiral couplings between the magnetic structure and the vortex beam.

**IV. STEADY STATE MAGNETIC STRUCTURE**

In order to investigate the effect of the vortex-induced interaction, we numerically determine the steady-state magnetic order of the 1-D chiral helimagnet. By defining the normalized effective magnetic field as \( \mathbf{h}_i \equiv -1 / J S^2 \left( \delta (\mathcal{H}_{1D}) / \delta \mathbf{n}_i \right) \), ...
the Landau–Lifshitz–Gilbert (LLG) equation becomes
\[ \frac{d\mathbf{n}_i}{d\tau} = -\frac{1}{1 + \alpha^2} \left[ \mathbf{n}_i \times \mathbf{h}_i + \alpha \mathbf{n}_i \times (\mathbf{n}_i \times \mathbf{h}_i) \right], \] (16)

where \( \alpha \) is the Gilbert damping constant. We use the dimensionless time \( \tau \) normalized by \( \hbar/(JS) \).

We show in figures 3 the modulation of the steady-state magnetization along the helical axis (z-axis) under the optical vortex radiation with \((l, s) = (1, 1)\). In the numerical simulation, we choose the following parameters: the lattice constant \( a = 1.212 \text{ nm} \) \([57]\), \( D/J = 0.16 \) \([38]\), \( \alpha = 0.3 \), \( \lambda = 1240 \text{ nm} \), the wavelength \( \lambda = 2\pi/k = 1240 \text{ nm} \), and \( B_0 = 10D/J \). We write the local spin direction as \( \mathbf{n}_i = (1, \theta(z_i), \phi(z_i)) \), where \( \theta \) is the polar angle and \( \phi \) is the azimuth angle with respect to the z-axis (see figure 3(a)). When the optical vortex field is absent, the angle \( \theta \) is uniformly 90° with \( \phi \) rotating around the z-axis. In figure 3(b), the deviation of the polar angle from 90° is plotted along the helical axis (z-axis). If the optical vortex field is absent, the angle \( \theta \) is uniformly 90° with \( \phi \) rotating around the z-axis. In figure 3(b), the deviation of the polar angle from 90° is plotted along the helical axis (z-axis). In figure 3(c), we show the difference between \( \Delta \theta^{(1,1)} \) and \( \Delta \theta^{(-1, -1)} \). One can see that \( \Delta \theta^{(1,1)} \) oscillates with the spiral pitch of the original chiral magnetic order. For the optical vortex radiation with \((l, s) = (-1, -1)\), the deviation \( \Delta \theta^{(-1, -1)} \) exhibits the oscillation with the same period as \( \Delta \theta^{(1,1)} \). In figure 3(c), we show the difference between \( \Delta \theta^{(1,1)} \) and \( \Delta \theta^{(-1, -1)} \). One can see that the envelop of the difference reflects the oscillation of \( b_1^{(1,1)} - b_1^{(-1, -1)} \). These results clearly show that the coupling between the LG beam and the spins can modulate the rigid chiral structure with the spatial structure of light. Such a spiral-pitch modulation of the macroscopic magnetic order in the helical axis is difficult to achieve by applying the external magnetic field. The results imply the possibility of the light with finite OAM to control the magnetic order.

In our simulation, we have assumed that the helical magnetic structure and the optical vortex are coaxial. The optical selection rule by the optical vortex is still valid even though the target ion does not lie at the vortex center. [12]
FIG. 4. (a) Dependence of the polar angle deviation $\Delta \theta$ on the intensity of the vortex-spin coupling $B_0 = 10^m D/J \ (m = -2, -1, 0, 1)$. The other parameters are the same in figure 3(b). (b) Dependence of $\Delta \theta$ on the Gilbert damping constant $\alpha = 0.032, 0.1, 0.3, 0.5, 0.8, 1$ with the other parameters set to be the same in figure 3(b).

Thus, our result is robust against the slight axis misalignment. In addition, when the light is obliquely incident with angle $\beta$, the vortex-induced coupling becomes $\mathbf{b}^i_{(\pm1, \pm1)} = -\cos \left[k \left(z_i + a/2\right)\hat{e}^x \pm \sin \left[k \left(z_i + a/2\right)\cos \beta \hat{e}^y \pm \sin \left[k \left(z_i + a/2\right)\sin \beta \hat{e}^z\right]\right]$. Although the coupling is reduced depending on the angle of incidence, it is possible to confirm the change in the magnetic structure accordingly. Besides, the $z$-component of $\mathbf{b}^i_{(\pm1, \pm1)}$ changes the Heisenberg exchange interaction so little that it can be ignored.

In the present setup, the helimagnet lies at the center axis of the optical vortex. Because the electric field intensity becomes weak at the vortex center, the vortex-electron coupling is considerably small. However, in the ion trap system, the excitation reflecting the transition selection rule is confirmed even near the center of the vortex axis [12]. We think that the results obtained here can be verified in this system by controlling the shape and intensity of the optical vortex beam. The coupling strength depends on the beam waist of the vortex beam. For the optical vortex with $l = s = \pm 1$, the optical transition matrix is approximately inversely proportional to the waist (see the Supplementary Material). Thus, in our calculation, we consider the vortex beam waist narrowed almost to the diffraction limit. On the other hand, in ion trap systems, optical transitions that reflect the OAM of light have been confirmed even when the ion position is shifted from the center of the vortex [14]. For the derivation of the optical vortex-induced interaction, it is sufficient to have the optical selection rule modified by the OAM of light. Thus, even if the 1D helimagnet is arranged off the central axis of the vortex, our results will not be significantly different. As long as the material absorbs the light with OAM, the various magnetic structure mediated by the itinerant electrons can be controlled.

There is one more thing to note about the system parameters to verify our work. In general, a strongly-driven system with an ac field such as lasers may increase the system temperature via coupling to its environment. As is mentioned in section III, the vortex-induced interaction essentially requires the finite absorption line width $\delta$. Then, the interaction should be affected by this heating effect to some extent. Thus, in this work, we have considered the parameter regime where the optical vortex-induced coupling is sufficiently smaller than the DM interaction, and is treated sufficiently within the perturbation regime. Seeing equations (12) and (15), one may be concerned that the third-order perturbation looks unreasonably large due to the factor $\delta^{-1}$. However, in fact, even when irradiating the strongest optical vortex beam among the parameters assumed here ($B_0 = 10^0 D/J$), the magnitude of the coefficient of the third term on the right-hand side of equation (13) is at most 0.01 that of the second term (DM interaction term). Here, the relaxation time is determined by the phenomenological line width $\delta$ for the optical absorption. To evaluate the heating effect in more detail, we need to microscopically take into account the coupling with a heat bath. There are many approaches that treat the dissipation effects in driving systems, e.g., Green’s function method [39–42] and quantum master equation method [34, 43–47].

V. CONCLUSION

We have formulated the spin-spin interaction induced by twisted light, i.e., optical vortex, in a 1-D chiral helimagnet such as chromium-intercalated niobium disulfide CrNb$_3$S$_6$. We have also shown by the LLG numerical calculation that such an optical vortex-induced interaction can modulate the chiral magnetic order to the unconventional phase structure.

Switchable control of magnetic orders is a fundamental and critical element for the evolution of spintronic de-
There are various attempts to control one- or two-dimensional chiral magnetic domain structure with cavity optomechanical system, electric currents, and electromagnetic fields. Here, we have microscopically derived a new type of spin-spin interaction induced by the optical vortex absorption. The vortex-induced interaction enables us to control the 1-D chiral spin order due to a completely different way. This interaction is attributed to the optical transition process between the d-orbitals, which requires the presence of the OAM of light. In addition, the spacial structure of the optical vortex beam provides a kind of chiral couplings with the magnetic structure. There are many degrees of freedom for optical vortex parameters, e.g., intensity, beam radius, polarization, and OAM, and these degrees of freedom can be helpful to such a control scheme. For instance, spatially-confined light can modulate magnetic structures locally. The local vortex-induced modulation is expected to affect the transport property of the quasiparticles, such as spin waves. The accessibility to the spin-wave dynamics in the chiral helimagnet has a possibility to open a route for new spintronic functionalities in device applications.

True completion of this study can present a principle for magneto-optics in a broader framework. Therefore, by clarifying the optical response of the magnetic order theoretically and suggesting the potential to control the magnetic order with light into experimental studies, we can blaze a trail in the field of opto-spintronics research.

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Supplementary Material: Twisted light-induced spin-spin interaction in a chiral helimagnet

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In this Supplementary Material, we show the derivation of the effective Hamiltonian (equation (13)), which is shown in section III of the main text. We also detail the optical transitions by the optical vortex shown in section IV.

I. EFFECTIVE SPIN-SPIN HAMILTONIAN

We show the derivation of the effective spin-spin Hamiltonian for the localized spin by the perturbative calculation, from tight-binding model Hamiltonian including the interaction among the \(d\)-electron, local moments, and optical vortex radiation.

A. Tight-binding model Hamiltonian

We employ a tight-binding model Hamiltonian in the whole system as for the electronic spin system in the chiral helimagnet and introduce the vector potential through the on-site interband excitation. The Hamiltonian includes the \(d\)-electrons energy, Hund’s coupling, and hopping energy between adjacent sites as follows:

\[
\mathcal{H}_{\text{ch}} = \mathcal{H}_0 + \mathcal{H}_{\text{hop}} + \mathcal{H}_{\text{SO}},
\]

where we define

\[
\mathcal{H}_0 = \sum_{i,\mu,\gamma} E_\mu c^\dagger_{i\mu\gamma} c_{i\mu\gamma} - J_H \sum_{i,\mu,\gamma,\gamma'} S_i \cdot s_i,
\]

\[
\mathcal{H}_{\text{hop}} = -\Gamma \sum_{i,j,\mu,\gamma,\gamma'} c^\dagger_{i\mu\gamma} c_{j\mu\gamma'},
\]

\[
\mathcal{H}_{\text{SO}} = \lambda \sum_i L_i \cdot s_i,
\]

with the energy of \(d\)-electrons \(E_\mu\), the annihilation operator \(c_{i\mu\gamma}\) such that the field operator of the electron is \(\psi_\gamma (r) = \sum_i \phi_i (r) c_{i\mu\gamma}\), where \(\phi_i (r)\) are the Wannier function centered on the site \(R_i\). The indices \(\{\mu,\nu\}\) and \(\{\gamma,\gamma'\}\) indicate the orbit and spin. Here we consider \(a_1\)- and \(e\)-symmetry band for the orbit. The constants \(J_H\), \(\Gamma\) and \(\lambda\) denote the Hund’s coupling energy between the electron spin \(s_i\) and the localized spin \(S_i\) with \(S = |S_i| = 3/2\), the hopping energy, and the spin-orbit coupling, respectively. We have neglected the spin-orbit interaction between the local momenta \(L_i \cdot S_i\). It contributes to the interband transition with spin-flipping whose transition energy is much larger than that by \(L_i \cdot s_i\) within the Hund’s rule. In addition, considering the effect of the optical vortex absorption, the \(L_i \cdot S_i\) term should be higher-order process.

In the absence of the spin-orbit interaction \(\mathcal{H}_{\text{SO}}\) and assuming the condition \(\Gamma \ll J_H\), the hopping \(\mathcal{H}_{\text{hop}}\) can be represented by employing the projection method as

\[
\mathcal{H}_{\text{hop}} = -\Gamma \sum_{i,j,\mu,\gamma,\gamma'} c^\dagger_{i\mu\gamma} \langle \gamma | P_i^0 P_j^0 | \gamma' \rangle c_{j\mu\gamma'},
\]

where \(P_i^0\) is the projection operator at the \(i\) site;

\[
P_i^0 = \frac{S_i \cdot \sigma + (S + 1) \sigma_0}{2S + 1}.
\]

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Here \( \mu \) denotes the orbital state with the lowest possible energy with the Hund’s coupling also considered. The Pauli matrix denotes the spin of itinerant electrons.

To introduce the interaction with the optical vortex, we introduce the ‘vector potential’ in the presence of the spin-orbit interaction \( \mathbf{A} \): 
\[
\chi^{(l,s)}(r, t) = -e A^{(l,s)}(r, t) - \left( \mu_B / 2e^2 \right) \sigma \times \mathbf{E}^{(l,s)}(r, t),
\]
with \( \sigma \) being the Pauli matrix. Here, \( \mu_B \) is the Bohr magneton, and \( \mathbf{E}^{(l,s)} \equiv i \omega \mathbf{A}^{(l,s)} - \nabla \varphi^{(l,s)} \). We additionally introduce the vector potential through the on-site interband excitation by the optical vortex radiation, i.e., \( \mathcal{H}'(t) = \mathcal{H}_{\text{ch}} + \mathcal{H}^{X}(t, t) \), where
\[
\mathcal{H}^{X}(r, t) = \frac{1}{2m} \sum_{i,\mu \neq \nu, \gamma, \gamma'} (i \mu \gamma)(p \cdot \chi^{(l,s)}(r, t) + \chi^{(l,s)}(r, t) \cdot \mathbf{p} + \text{H.c.}) |i\nu\gamma\rangle c_{i\mu\gamma}^\dagger c_{i\nu\gamma'},
\]
with \( m, p \) and \( E_{\mu\nu} \) being the mass, momentum of electron, and transition energy. In addition, we define the matrix element for some operator \( \mathcal{O} \) as 
\[
\langle i\mu\gamma | \mathcal{O} | i\nu\gamma' \rangle = \langle \gamma | \int d\mathbf{r} \phi_{i\mu}^\dagger(\mathbf{r}) \mathcal{O}(\mathbf{r}) \phi_{i\nu}(\mathbf{r}) |\gamma' \rangle.
\]
The electric field is assumed to be resonant with \( E_g \), where \( E_g = E_e - E_{a_1} + SJ_H \) is the gap energy including the Hund’s coupling.

### B. Treatment of the perturbative Hamiltonian

We derive the effective model considering the perturbations with respect to the hopping \( \mathcal{H}_{\text{hop}} \), spin-orbit interaction \( \mathcal{H}_{\text{SO}} \), and optical vortex absorption \( \mathcal{H}^{X}(t) \). The unperturbed Hamiltonian is \( \mathcal{H}_0 \), and the itinerant electrons in the lower band \( (a_1) \) have their spins parallel to the localized spins in the unperturbed ground state. Thus, we impose the constraint not to break the Hund rule throughout the perturbation calculation. Here we consider the time evolution operator in the interaction picture
\[
U(t, 0) = \mathcal{T} \exp \left\{ -i \frac{\hbar}{\hbar} \int_0^t (\mathcal{H}_{\text{hop}} + \mathcal{H}_{\text{SO}} + \mathcal{H}^{X}(t')) dt' \right\},
\]
where \( \mathcal{T} \) is the time-ordering operator. The zero-th order \( U^{(0)}_1(t; 0) \) is identity. We consider only the energy terms \( \mathcal{E}^{(i)} \) from one to three in \( \zeta \) are treated as the \( \zeta \)-th order perturbation.

**First-order perturbation** The first-order term is just \( \mathcal{H}_{\text{hop}} \), then
\[
\mathcal{H}^{(1)} = -\Gamma \sum_{i,\mu, \nu, \gamma, \gamma'} \chi_{i\mu\gamma} \langle \gamma | P_i^0 P_j^0 | \gamma' \rangle c_{i\mu\gamma}^\dagger c_{i\nu\gamma'}.
\]
The term indicates that the localized spins are aligned for itinerant electrons to have more extended wave forms. Here we assume \( S_j \) being a classical vector \( S(\cos \varphi \sin \vartheta \sin \varphi \sin \vartheta \cos \vartheta) \), then
\[
P_i^0 = \frac{1}{2S + 1} \begin{pmatrix} S \cos \vartheta + S + 1 & S \sin \vartheta e^{-i\varphi_j} \\ S \sin \vartheta e^{i\varphi_j} & -S \cos \vartheta + S + 1 \end{pmatrix}.
\]
Diagonalizing \( P_j \), we obtain the eigensystem,
\[
\tilde{P}_j = M_j^{-1} P_j^0 M_j = \begin{pmatrix} 1 & 0 \\ 0 & 1/(2S + 1) \end{pmatrix},
\]
and
\[
M_j = \begin{pmatrix} e^{-i\varphi_j} \cos \vartheta_j & e^{-i\varphi_j} \sin \vartheta_j \\ e^{i\varphi_j} \sin \vartheta_j & e^{i\varphi_j} \cos \vartheta_j \end{pmatrix}.
\]
Thus, the Hamiltonian can be written by those matrices
\[
\mathcal{H}^{(1)} = -\Gamma \sum_{i,\mu, \nu, \gamma, \gamma'} \chi_{i\mu\gamma} \langle \gamma | M_j \tilde{P}_j M_i^{-1} M_j \tilde{P}_j M_j^{-1} | \gamma' \rangle c_{i\mu\gamma}^\dagger c_{i\nu\gamma'}.
\]
Here we consider the low energy state only, and neglect the higher inhomogeneous hopping energy which may cause charge accumulation. The Hamiltonian reduces to

$$H^{(1)}(t) \approx -t \sum_{i,j,\mu} \left[ \cos \varphi_{ij} \cos \left( \frac{\vartheta_{ij}}{2} \right) \right] d_{ij\mu}^+ d_{i\mu-} + \text{H.c.},$$

where $\varphi_{ij} = \varphi_i - \varphi_j$, $\vartheta_{ij} = \vartheta_i - \vartheta_j$ and

$$d_{i\mu-} = e^{i \varphi_{ij} / 2} c_{j\mu +} + e^{-i \varphi_{ij} / 2} \sin \vartheta_{ij} c_{j\mu i},$$

is the field operator of the electrons in the eigenstate of lower energy. Since the corrected hopping energy is proportional to $\cos (\vartheta_{ij}/2)$ for the lower energy, we can reach the equation [S9], which favors the ferromagnetic state.

**Second-order perturbation** For the second-order term, it depends on the transition matrix of two products from each of the hopping $H_{\text{hop}}$, spin-orbit interaction $H_{\text{SO}}$, and optical vortex absorption $H_{\text{X}}(t)$. The only ones for which transitions that exist are the following;

$$\langle i, \mu, \gamma | U_1^2(t; 0) | j, \mu, \gamma' \rangle$$

$$= -\frac{1}{\hbar^2} \int_0^t dt_1 \int_0^{t_1} dt_2 \left( \langle i, \mu, \gamma | H_{\text{hop}}(t_1) H_{\text{SO}, 1} H_{\text{SO}, 1} (t_2) | j, \mu, \gamma' \rangle + \langle i, \mu, \gamma | H_{\text{SO}, 1} (t_1) H_{\text{hop}, 1} (t_2) | j, \mu, \gamma' \rangle \right)$$

$$- \frac{1}{\hbar^2} \int_0^t dt_1 \int_0^{t_1} dt_2 \left( \langle i, \mu, \gamma | H_{\text{SO}, 1} (t_1) \tilde{H}_X^2 (t_2) | j, \mu, \gamma' \rangle + \langle i, \mu, \gamma | \tilde{H}_X (t_1) H_{\text{hop}, 1} (t_2) | j, \mu, \gamma' \rangle \right)$$

$$= \frac{i}{\hbar} \left[ \frac{E_g}{E_g - \hbar \omega - i \delta} \left( \frac{e^{i E_g \omega \delta} / h - 1}{E_g + \hbar \omega + i \delta} \right) + \frac{E_g}{E_g + \hbar \omega - i \delta} \left( \frac{e^{i E_g \omega \delta} / h - 1}{E_g - \hbar \omega + i \delta} \right) \right] \times \left( 1 - \delta_{ij} \right) \left( 1 - \delta_{\mu \delta} \right) \sum_{\gamma''} H_{\text{hop}}^2 (i \mu \gamma ; j \nu \gamma'' ; j \nu \gamma')$$

$$- \frac{1}{\hbar E_g} \left[ \frac{E_g}{E_g - \hbar \omega + i \delta} \left( \frac{e^{i E_g \omega \delta} / h - 1}{E_g + \hbar \omega + i \delta} \right) + \frac{E_g}{E_g - \hbar \omega - i \delta} \left( \frac{e^{i E_g \omega \delta} / h - 1}{E_g + \hbar \omega - i \delta} \right) \right] \times \left( 1 - \delta_{ij} \right) \left( 1 - \delta_{\mu \delta} \right) \sum_{\gamma''} H_{\text{dip}}^2 (i \mu \gamma ; i \nu \gamma'' ; j \nu \gamma')$$

where

$$\mathcal{H}_{\text{hop}} = -\Gamma \sum_{i,j,\mu,\nu,\gamma,\gamma'} (1 - \delta_{ij}) c_{i \mu \gamma}^d \langle \gamma | P_{\mu \nu}^0 P_{\nu \gamma}^0 | \gamma' \rangle c_{j \nu \gamma'},$$

and the transition amplitude

$$H_{\text{hop}}^2 (i \mu \gamma ; j \nu \gamma') = \langle i, \mu, \gamma | H_{\text{hop}}(t) | j, \nu, \gamma' \rangle,$$

$$H_{\text{SO}} (i \mu \gamma ; i \nu \gamma') = \langle i \mu \gamma | H_{\text{SO}} | i \nu \gamma' \rangle,$$

$$H_{\text{dip}}^2 (i \mu \gamma ; i \nu \gamma') = -\frac{i \hbar}{E_g} \langle i \mu \gamma | H_{\text{X}}(t) | i \nu \gamma' \rangle,$$

with $\delta$ being the phenomenological line width for the optical transition. Here $H_{\text{X}}(t)$ is $H_{\text{X}}(r, t)$ with the temporal dependence omitted. We relax the restriction of the band transition we put on $H_{\text{hop}}$ in equation [S3] because other orbitals can be accessed via the spin-orbit interaction. Assuming that the perturbation energy is much smaller than the bandgap energy, we use secular approximation and ignore the temporal oscillation term like the Rabi oscillation under the resonant condition [S3, S4]. In addition, the period of the neglected oscillation $\sim 2 \pi h / E_g$ is much shorter than the time step of the subsequent simulation. Such a oscillation should be averaged out during the step time. It thus becomes

$$\langle i, \mu, \gamma | U_1^2(t; 0) | j, \mu, \gamma' \rangle \approx -\frac{i}{\hbar E_g} \left( 1 - \delta_{ij} \right) \left( 1 - \delta_{\mu \delta} \right) \sum_{\gamma''} \left( H_{\text{hop}}^2 (i \mu \gamma ; j \nu \gamma'' ; j \nu \gamma') + H_{\text{SO}} (i \mu \gamma ; i \nu \gamma'' ; j \nu \gamma') + H_{\text{dip}}^2 (i \nu \gamma'' ; j \nu \gamma') \right).$$
Since \( \langle \mu | \mathbf{L}_i | \nu \rangle \) (\( \mu \neq \nu \)) is purely imaginary, we obtain the second-order perturbation energy as

\[
\mathcal{E}^{(2)} = \sum_{i,j,\mu,\nu,\gamma,\gamma'} \frac{1}{E_g} (1 - \delta_{ij}) (1 - \delta_{\mu\nu}) \left[ H_{\text{hop}}^{(i\mu\gamma; j\nu\gamma')} H_{\text{SO}}^{(j\nu\gamma'; j\mu\gamma)} + H_{\text{SO}}^{(i\mu\gamma; i\nu\gamma')} H_{\text{hop}}^{(i\nu\gamma'; j\mu\gamma)} \right].
\]

We substitute \( H_{\text{hop}}^{(i\mu\gamma; j\nu\gamma')} \) and \( H_{\text{SO}}^{(i\mu\gamma; i\nu\gamma')} \), and simplify the terms by using the commutator. Thus, it can be rewritten as the perturbation Hamiltonian in the second order [S3]:

\[
\mathcal{H}^{(2)} = \frac{\Gamma}{E_g} \sum_{i,j,\mu,\nu,\gamma,\gamma'} (1 - \delta_{ij}) (1 - \delta_{\mu\nu}) c_{i\mu\gamma}^\dagger \langle \gamma | \left( \langle a_1 | \mathbf{L}_i | e \rangle \cdot P_{\sigma_j}^{(i)} \right) | \gamma \rangle c_{j\nu\gamma},
\]

where

\[
\mathbf{v}_i = -\frac{i\lambda}{E_g} \langle a_1 | \mathbf{L}_i | e \rangle \times \mathbf{S}_i.
\]

The vector [S11] corresponds to equation (11) in the main text.

Third-order perturbation For the third-order term, the transitions that exist are for the following:

\[
(i, \mu, \gamma | U_1^{(3)} (t; 0) | j, \mu, \gamma' \rangle = + \frac{i}{\hbar} \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_2} dt_3 \langle i, \mu, \gamma | \mathcal{H}_{\text{hop}}, 1 (t_1) \mathcal{H}_{\text{SO}}, 1 (t_2) \mathcal{H}_X (t_3) | j, \mu, \gamma' \rangle
\]

The use of the secular approximation under the resonant condition as the same way of the second-order perturbation. In addition, we consider the steady state where \( e^{-\delta t} \sim 0 \) with the line width \( \delta \). For instance, one can find

\[
i \int_a^b dt_1 \int_0^{t_1} dt_2 \int_0^{t_2} dt_3 \langle i, \mu, \gamma | \mathcal{H}_{\text{hop}}, 1 (t_1) \mathcal{H}_X (t_2) \mathcal{H}_{\text{SO}}, 1 (t_3) | j, \mu, \gamma' \rangle
\]

\[
= i \frac{e^{i(h\omega+i\delta)t/b}}{(h\omega+i\delta)^2} - \frac{it/h}{(h\omega+i\delta)} - \frac{e^{i(E_g+h\omega+i\delta)t/b}}{(E_g+h\omega+i\delta)^2} + \frac{it/h}{(E_g+h\omega+i\delta)}
\]

\[
\times (1 - \delta_{ij}) (1 - \delta_{\mu\nu}) \sum_{\gamma'' \gamma'''} H_{\text{hop}}^{(i\mu\gamma; j\nu\gamma''')} H_{\text{SO}}^{X \text{dip} (j\nu\gamma'''; j\mu\gamma')} H_{\text{SO}}^{(j\mu\gamma'''; j\nu\gamma''')} H_{\text{SO}}^{(j\nu\gamma''''; j\mu\gamma')}
\]

\[
\approx \frac{it}{\hbar^2 \delta} (1 - \delta_{ij}) (1 - \delta_{\mu\nu}) \sum_{\gamma'' \gamma'''} H_{\text{hop}}^{(i\mu\gamma; j\nu\gamma''')} H_{\text{SO}}^{X \text{dip} (j\nu\gamma'''; j\mu\gamma')} H_{\text{SO}}^{(j\mu\gamma'''; j\nu\gamma''')} H_{\text{SO}}^{(j\nu\gamma''''; j\mu\gamma')}.
\]
The other terms reduce to zero or time-independent constant. Thus, we obtain the third-order perturbation energy

$$
E^{(3)} = -\frac{1}{\hbar\delta} \sum_{i,j,m,n,\gamma,\gamma'} (1 - \delta_{ij})(1 - \delta_{\mu\nu}) \sum_{\gamma''} \left[ H_{\text{hop}}(i\mu\gamma'; j\mu'\gamma'') H_{\text{dip}}^x(i\nu\gamma''; j\nu'\gamma''') H_{SO}(i\nu'\gamma'''; j\nu''\gamma') + H_{\text{dip}}(i\mu\gamma; j\mu'\gamma') H_{\text{dip}}^x(i\nu\gamma''; j\nu'\gamma'') H_{SO}(i\nu'\gamma'''; j\nu''\gamma') + H_{\text{hop}}(i\mu\gamma; j\nu\gamma'') H_{\text{hop}}(i\nu'\gamma'''; j\nu''\gamma') H_{SO}(i\nu''\gamma'''; j\nu''\gamma') + H_{\text{dip}}(i\mu\gamma; j\nu\gamma'') H_{\text{dip}}(i\nu'\gamma'''; j\nu''\gamma') H_{SO}(i\nu''\gamma'''; j\nu''\gamma') \right].
$$

To symmetrize Hamiltonian for the site indices, we take the additional terms which are zero. It can be rewritten as the third-order perturbation Hamiltonian, substituting $H_{\text{hop}}(i\mu\gamma; j\mu'\gamma')$, $H_{SO}(i\nu; i\nu')$ and $H_{\text{dip}}^x(i\nu; j\nu')$;

$$
H^{(3)} = \frac{\Gamma}{2\hbar\delta} \sum_{i,j,m,n,\gamma,\gamma'} c_{i\mu\gamma}^\dagger (1 - \delta_{ij})(1 - \delta_{\mu\nu}) \left[ \left( a_{i1}|L_i|e\right) \left( a_{i1}|r_i \cdot \chi_i^{(s)}|e\right), P^0 \right] \sigma_i P_j^0 + \left( |e| L_j|a1\right) \left( |e| r_j \cdot \chi_j^{(s)}|a1\right), P^0 \right] \sigma_i P_j^0 + \left( a_{i1}|L_j|e\right) \left( a_{i1}|r_j \cdot \chi_j^{(s)}|e\right), P_j^0 \sigma_j + \left( |e| L_j|a1\right) \left( |e| r_j \cdot \chi_j^{(s)}|a1\right), P_j^0 \sigma_j \right] \gamma^\prime \rangle e_{j\nu\gamma'} \left[ + P_j^0 \left( \text{Im} \left( a_{i1}|B_j^{(s)}|e\right) \cdot \left( a_{i1}|L_j|e\right) \times S_j\right) \sigma_0 + \left( \text{Im} \left( a_{i1}|B_j^{(s)}|e\right) \times \left( a_{i1}|L_j|e\right) \times S_j\right) \sigma_0 P_j^0 \right] \gamma^\prime \rangle \left( e_{j\mu\nu} \right),
$$

where

$$
B_j^{(s)} = -\frac{\mu B}{2c^2} E_j^{(s)} \times r_j,
$$

and

$$
v'_i = \frac{2i\lambda}{\hbar\delta} \left( a_{i1}|L_i|e\right) \times \left( \text{Im} \left( a_{i1}|B_i^{(s)}|e\right) \times S_i\right).
$$

The vector $S_{13}$ corresponds to equation (12) in the main text. Since the last term in equation (S12) does not related with the itinerant spin $\sigma$, it does not affect the localized spins at all. The terms including ‘vector potential’ $-eA_j^{(s)}$ in $\chi_j^{(s)}$ vanish in the effective energy for the same reason. Besides, even within the secular approximation, the term with two optical vortex terms also exists as a perturbation. However, the magnitude of the perturbation term contributed by the optical vortex is small. In this study, we ignore the perturbation terms that contain more than two optical vortex absorption terms. Thus, we redefine $H^{(3)}$ as

$$
H^{(3)} = -\frac{\Gamma}{2S+1} \sum_{i,j,m,n,\gamma,\gamma'} c_{i\mu\gamma}^\dagger (1 - \delta_{ij})(1 - \delta_{\mu\nu}) \left[ \left( P_j^0 \sigma_j \cdot v'_i + |e| \cdot \sigma_i P_j^0 \right) \gamma^\prime \right] e_{j\mu\gamma'}.
$$
C. Effective spin-spin interaction Hamiltonian

Defining the whole Hamiltonian $H'$ by summing $H^{(1)}, H^{(2)}$ and $H^{(3)}$ for the total perturbation Hamiltonian, it becomes

$$H' = H_0 - \Gamma \sum_{i,j,\mu,\nu,\gamma,\gamma'} c_{i\mu\gamma}^\dagger (1 - \delta_{ij}) (1 - \delta_{\mu\nu}) \langle \gamma | P_i P_j | \gamma' \rangle c_{j\nu\gamma'},$$

(S14)

where we define the projection operator as

$$P_i = \frac{(S_i + v_i + v'_i) \cdot \sigma + (S + 1) \sigma_0}{2S + 1}.$$  

The three mutually orthogonal vectors $S_i$, $v_i$, and $v'_i$ incorporate the effects of the first-, the second-, and the third-order perturbation, respectively. The Hamiltonian (S14) is just the one in equation (9) of the main text.

As is the derivation from (S6) to (S9), we derive the effective Hamiltonian from $H'$ by substituting $\vartheta_i$ and $\varphi_i$ with $\vartheta_i + \Delta \vartheta_i$ and $\varphi_i + \Delta \varphi_i$ which are the angular coordinates of the vector $S_i + v_i + v'_i$. The resultant spin-spin interactions become (S15)

$$\cos \left( \frac{\vartheta_{ij} + \Delta \vartheta_{ij}}{2} \right) \approx J (S_i + v_i + v'_i) \cdot (S_j + v_j + v'_j) = J S_i \cdot S_j - D_{ij} \cdot (S_i \times S_j) + S_i^\dagger \left( D_{ij} E_{ij}^{(l,s)} + B_{ij}^{(l,s)} D_{ij}^\dagger \right) S_j,$$

where

$$D_{ij} = \frac{\lambda J}{E_g} \text{Im} \left( \langle L_i \rangle_{\mu\nu} - \langle L_j \rangle_{\mu\nu} \right),$$

$$B_{ij}^{(l,s)} = \frac{2E_g}{\hbar} \text{Im} \left( \langle B_{ij}^{(l,s)} \rangle_{\mu\nu} - \langle D_j^{(l,s)} \rangle_{\mu\nu} \right).$$

Here we set the magnitude of the Heisenberg coupling interaction as $J$.

II. OPTICAL TRANSITION

We show the theoretical form for the optical transition by the optical orbital angular momentum of the optical vortex. For the 3$d$-orbitals of Cr: $e$, $a_1$, and $e'$ orbitals, we consider the interband transition of the $d$-electron by optical vortices via the dipole interaction. The transition strength from $Y_{2,0}$ to $Y_{2,m}$ ($m = \pm 1, \pm 2$) describes the following integration,

$$\int drr^2 |R_{32}|^2 \int d\vartheta d\varphi \sin \theta \left[ \begin{array}{c} Y_{2,+2}^2 \\ Y_{2,-2}^2 \\ Y_{2,+1}^{2} \\ Y_{2,-1}^{2} \end{array} \right] r \cdot (\sigma \times E^{(l,s)}) \cdot Y_{2,0},$$

where the radial function

$$R_{32} (r) = \frac{4}{27\sqrt{10}} \left( \frac{1}{3r_0} \right)^\frac{3}{2} \left( \frac{r}{r_0} \right)^2 e^{-\frac{r}{b\sigma}},$$

with the Bohr radius $r_0$ and spherical harmonics

$$Y_{2,0} (\theta, \phi) = \sqrt{\frac{5}{16\pi}} (3 \cos^2 \theta - 1) ,$$

$$Y_{2,\pm 1} (\theta, \phi) = \pm \sqrt{\frac{15}{8\pi}} \sin \theta \cos \theta e^{\pm \imath \phi} ,$$

$$Y_{2,\pm 2} (\theta, \phi) = \sqrt{\frac{15}{32\pi}} \sin^2 \theta e^{\pm \imath \phi} .$$
For the transition strength, when \((l, s) = (+1, +1)\), it becomes

\[
\int dr r^2 |R_{32}|^2 \int d\theta d\phi \sin \theta Y_{2, +1} \cdot \left( \sigma \times E^{(l,s)} \right) Y_{2, 0} \approx \frac{1}{4} \omega A_0 r_0 \frac{96}{\pi} \sqrt{\frac{3}{5}} \left( \frac{r_0}{w_0} \right) \langle \sigma \rangle_{\gamma \gamma'} \cdot \begin{bmatrix} 1 \\ i \\ 0 \end{bmatrix},
\]

\[
\int dr r^2 |R_{32}|^2 \int d\theta d\phi \sin \theta Y_{2, +2} \cdot \left( \sigma \times E^{(l,s)} \right) Y_{2, 0} \approx \omega A_0 r_0 \frac{96}{\pi} \sqrt{\frac{3}{5}} \left( \frac{r_0}{w_0} \right) \langle \sigma \rangle_{\gamma \gamma'} \cdot \begin{bmatrix} 0 \\ 0 \\ -1 \end{bmatrix},
\]

\[
\int dr r^2 |R_{32}|^2 \int d\theta d\phi \sin \theta \left\{ Y_{2, -2}^* \right\} \cdot \left( \sigma \times E^{(l,s)} \right) Y_{2, 0} = 0.
\]

On the other hand, when \((l, s) = (-1, -1)\), one can see

\[
\int dr r^2 |R_{32}|^2 \int d\theta d\phi \sin \theta Y_{2, -1} \cdot \left( \sigma \times E^{(l,s)} \right) Y_{2, 0} \approx \frac{1}{4} \omega A_0 r_0 \frac{96}{\pi} \sqrt{\frac{3}{5}} \left( \frac{r_0}{w_0} \right) \langle \sigma \rangle_{\gamma \gamma'} \cdot \begin{bmatrix} 1 \\ -i \\ 0 \end{bmatrix},
\]

\[
\int dr r^2 |R_{32}|^2 \int d\theta d\phi \sin \theta Y_{2, -2} \cdot \left( \sigma \times E^{(l,s)} \right) Y_{2, 0} \approx \omega A_0 r_0 \frac{96}{\pi} \sqrt{\frac{3}{5}} \left( \frac{r_0}{w_0} \right) \langle \sigma \rangle_{\gamma \gamma'} \cdot \begin{bmatrix} 0 \\ 0 \\ 1 \end{bmatrix},
\]

\[
\int dr r^2 |R_{32}|^2 \int d\theta d\phi \sin \theta \left\{ Y_{2, +2}^* \right\} \cdot \left( \sigma \times E^{(l,s)} \right) Y_{2, 0} = 0.
\]

Even when \((l, s) = (+3, -1)\) or \((-3, +1)\), the transition strength is still finite, but its order is the same of or smaller than the one for \((l, s) = (\pm 1, \pm 1)\) multiplied by \((r_0/w_0)^2\).

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