Rapid Communications

Two-photon driven magnon-pair resonance as a signature of spin-nematic order

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We theoretically study the nonlinear magnetic resonance driven by intense lasers or electromagnetic waves in a fully polarized frustrated magnet near a less-visible spin-nematic ordered phase. In general, both magnons and magnon pairs (two-magnon bound state) appear as the low-energy excitation in the saturated state of spin-nematic magnets. Their excitation energies are usually in the range between 10 GHz and 10 THz. Magnon pairs with angular momentum $2\hbar$ can be excited by the simultaneous absorption of two photons, and such multiphoton processes occur if the applied THz laser is strong enough. We compute laser-driven magnetic dynamics of a frustrated four-spin system with both magnon ($\hbar$) and magnon-pair ($2\hbar$)-like excitations, which is analogous to a macroscopic frustrated magnet with a spin-nematic phase. We estimate the required strength of the magnetic field of a laser for the realization of two-photon absorption, taking into account dissipation effects with the Lindblad equation. We show that an intense THz laser with an ac magnetic field of 0.1–1.0 T is enough to observe magnon-pair resonance.

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Introduction. Laser science and technology have progressed in the last decades, and have stimulated the study of condensed matter and nonequilibrium physics because the progress makes it possible to observe or create a variety of excitations in solids, liquids, and so on. In recent years, the laser science in the range of 0.1–1 THz [1–4] has greatly developed and we can use THz-laser pulses with an intensity of 1 MV/cm ($\sim$0.3 T). As a result, it is becoming possible to control magnetic excitations or textures with a laser because the photon energy in the THz range is comparable to that of magnetic excitations, especially those of antiferromagnets [5]. Photoinduced magnetic phenomena have also been actively explored as issues of magneto-optics [6] and spintronics [7]. Several groups have observed linear and nonlinear magnetic responses for THz lasers or waves: for instance, large magnetic resonances driven by THz lasers or waves [8,9], high harmonic generation (HHG) induced by a THz-laser pulse in an antiferromagnetic insulator [10], electromagnon resonance driven by an electric field of THz waves [11–13], dichroisms in a ferrimagnet driven by THz vortex beams [14], etc. In addition, the electron spin resonance (ESR) driven by electromagnetic waves in the range between 10 GHz and 1.0 THz has long been studied [15–25]. Microscopic or quantum theories for magnetic dynamics driven by intense electromagnetic waves have also begun to develop: Floquet engineering in magnetic systems [26–30], control of exchange couplings in Mott insulators [31,32], ultrafast creation or control of magnetic defects in chiral magnets [33–36], applications of topological light waves [37,38], laser-driven spin current in magnetic insulators [39,40], HHG in quantum spin systems [41], etc.

Motivated by these activities, in this Rapid Communication, we theoretically consider how to detect a signature of less-visible spin-nematic (quadrupolar) order in magnetic insulators with a laser or electromagnetic wave. A spin-nematic ordered phase [42–44] is a physical state with a spin quadrupolar order and without any spin dipolar (magnetic) order. Its order parameter is defined as an expectation value of the tensor product of two spins. In the present work, we focus on the spin-nematic order in S^x-S^y plane defined by $\langle S_r^+S_{r'}^+ + S_r^-S_{r'}^-\rangle$, which accompanies the breaking of U(1) spin rotation symmetry around the S^z axis. It is important to inhibit the usual spin order for the emergence of such spin-nematic states and thereby frustrated magnets often become a good candidate. Another point is that not only standard magnons but also magnon pairs (molecules of two magnons) [45–47] usually appear in field-induced fully polarized (i.e., ferromagnetic) states of spin-nematic magnets [see Fig. 1(a) and the Supplemental Material [48]]. If the applied magnetic field is decreased and the magnon-pair band becomes lower than the energy of the saturated state, the Bose-Einstein condensation (BEC) of magnon pairs occurs. The product of neighboring spins $S_r^-S_{r'}^-$ ($S_r^+S_{r'}^+$) can be viewed as the creation (annihilation) operator of a magnon pair. In their BEC state,

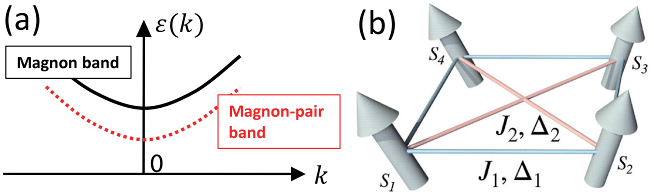

FIG. 1. (a) Generic band structures of a magnon and magnon pair in the field-driven fully polarized state of a frustrated magnet near a spin-nematic phase [48]. (b) Frustrated four-spin model [see Eq. (1)].

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these operators have finite expectation values $\langle S_r^\pm S_{r'}^\pm \rangle \neq 0$ and therefore it means the emergence of a spin-nematic order. This is a typical scenario for generating a spin-nematic order.

Generally, it is quite difficult to detect clear evidence of the spin-nematic order compared to usual magnetic orders because its detection requires a direct observation of a tensor product of two spins $\langle S_r^+ S_{r'}^+ + S_r^- S_{r'}^- \rangle$ or a four-point spin-nematic correlation function such as $\langle S_r^+ S_{r+\delta}^+ S_0^- S_\delta^- \rangle$. For a spin-nematic quasi-long-range ordered phase in one-dimensional magnets, it has been shown [49,50] that NMR [51–55], neutron scattering spectra [56,57], and the spin Seebeck effect [58] are very useful to detect its signature, while clear experimental ways of detecting spin-nematic long-range orders have not been well established [59–61]. On the other hand, as we mentioned above, magnon-pair excitations almost always appear in the saturated state of spin-nematic magnets including both spin-nematic long-range and quasi-long-range ordered phases. We here discuss a method of observing magnon pairs with intense laser or electromagnetic waves as a way of obtaining indirect but strong evidence of spin-nematic orders. Magnons and photons carry an angular momentum $\hbar$, while magnon pairs have an angular momentum $2\hbar$. Therefore, magnon pairs can be excited through two-photon absorption and such multiphoton processes can be realized with a sufficiently strong laser. We compute the time evolution of laser-driven spin dynamics in a frustrated nanospin model that is analogous to spin-nematic magnets. We take into account the dissipation effect, which is quite important to estimate realistic spectra of magnon-pair resonance, by applying a quantum master equation with the Lindblad approximation. We show that magnon-pair resonance spectra can be detected by currently available THz lasers or GHz waves.

Model and method. Here, we define our model for studying laser-driven spin dynamics. We focus on the frustrated four-spin model described by Fig. 1(b). The Hamiltonian is given by

$$\mathcal{H}_0 = \sum_{j=1-4} \left(J_1 \boldsymbol{S}_j \cdot \boldsymbol{S}_{j+1} + \Delta_1 S_j^z S_{j+1}^z\right) - H S_{\rm tot}^z + \sum_{j=1,2} \left(J_2 \boldsymbol{S}_j \cdot \boldsymbol{S}_{j+2} + \Delta_2 S_j^z S_{j+2}^z\right), \tag{1}$$

where $\boldsymbol{S}_j$ is the electron spin-$\frac{1}{2}$ operator on the jth site (j: mod 4), and $S_{\rm tot}^\alpha = \sum_{j=1}^4 S_j^\alpha$ is the sum of four spins. Here, $J_{1,2}$ are the competing exchange interactions, $\Delta_{1,2}$ are the Ising anisotropy constants, and $H = g\mu_B h_0$ is the strength of Zeeman coupling for an applied static magnetic field h_0 (g is the g-factor and μ_B is the Bohr magneton). This Rapid Communication uses the unit of $\hbar = 1$. The eigenenergies and normalized eigenstates for $\mathcal{H}_0$ are respectively described as $\{E_n\}$ and $\{|\psi_n\rangle\}$ with $E_1 \leqslant E_2 \leqslant \cdots \leqslant E_{16}$. It is shown that a two-dimensional system consisting of weakly coupled four-spin models (1) exhibits a spin-nematic order at $J_1/J_2 \sim -2$ [62]. The model (1) thereby may be regarded as an analog of a bulk spin-nematic magnet. Hereafter, we adopt $J_1 = -2.2$, $J_2 = 1$, and $\Delta_1 = \Delta_2 = 0.24$, in which the energy eigenstates are classified as a spin quintet $|S_{\rm tot}, S_{\rm tot}^z\rangle = |2, M\rangle$, three spin triplets $|1, M\rangle_p$, and two spin singlets $|0, 0\rangle_q$ ($p = 1, 2, 3$ and $q = 1, 2$). As we show later, a finite anisotropy $\Delta_{1,2}$ generates a difference between the resonant frequencies of a magnon and magnon pair. The details of $\{|\psi_n\rangle\}$ and $\{E_n\}$ are explained in the Supplemental Material [48].

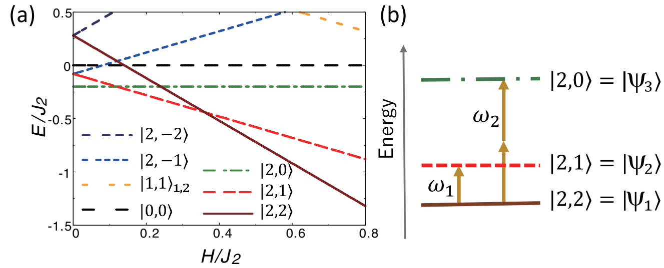

FIG. 2. (a) Field (H) dependence of energy levels $|S_{\rm tot}, M\rangle$ of the nanospin model (1) with $J_1 = -2.2$, $J_2 = 1$, and $\Delta_1 = \Delta_2 = 0.24$. (b) Low-energy level structure at a high field $H = 0.56$. $\omega_{1(2)}$ is the magnon (magnon-pair) resonance frequency. The unit of $\hbar = 1$ is used.

Figure 2(a) shows the field dependence of energy levels in the low-energy range. Since we consider the fully polarized state and magnon-pair-like excitations in the model (1), we set $H = 0.56$, in which the ground state is fully polarized ($S_{\rm tot}^z = 2$) as shown in Fig. 2(a). Then we calculate the spin dynamics under the application of an intense THz laser or electromagnetic wave. The ac Zeeman coupling is given by $\mathcal{H}_{\rm cp}(t) = \frac{A}{2}(e^{-i\omega t} S_{\rm tot}^+ + e^{+i\omega t} S_{\rm tot}^-)$ for a circularly polarized laser and $\mathcal{H}_{\rm lp}(t) = A\cos(\omega t) S_{\rm tot}^x = \frac{A}{2}\cos(\omega t)(S_{\rm tot}^+ + S_{\rm tot}^-)$ for a linearly polarized one. ω is the angular frequency of the laser, $A = g\mu_B h_{\rm ac}$ denotes the amplitude of the magnetic field of the laser, and $S_{\rm tot}^\pm = S_{\rm tot}^x \pm i S_{\rm tot}^y$. The ac electric field is negligible since the THz photon energy is usually much smaller than the charge gap of the magnets.

To see the time evolution of the nanomagnet with ac Zeeman coupling, we numerically solve the quantum master equation for density matrix $\rho(t)$ [63–66],

$$\dot{\rho}(t) = -i[\mathcal{H}(t), \rho(t)] + \sum_{j=2}^{16} [N(\omega_j) + 1]\left(L_j \rho(t) L_j^\dagger - \frac{1}{2}\{L_j^\dagger L_j, \rho(t)\}\right) + \sum_{j=2}^{16} N(\omega_j)\left(L_j^\dagger \rho(t) L_j - \frac{1}{2}\{L_j L_j^\dagger, \rho(t)\}\right), \tag{2}$$

with the fourth-order Runge-Kutta method. The first line on the right-hand side represents the dynamics driven by the Hamiltonian $\mathcal{H}(t) = \mathcal{H}_0 + \mathcal{H}_{\rm cp}(t)$ [or $\mathcal{H}_0 + \mathcal{H}_{\rm lp}(t)$], while the second and third lines give a so-called Lindblad-type dissipation. The Lindblad (jump) operator $L_j = \sqrt{\gamma}|\psi_{j-1}\rangle\langle\psi_j|$ and γ is the coupling constant between the system and environment. The value of $\hbar/\gamma$ is the typical time of relaxation. If there is a degeneracy $E_{j-1} = E_j$, we modify the jump operators as $L_{j-1} = \sqrt{\gamma/2}|\psi_{j-2}\rangle(\langle\psi_{j-1}| + \langle\psi_j|)$, $L_j = 0$, and $L_{j+1} = \sqrt{\gamma/2}(|\psi_{j-1}\rangle + |\psi_j\rangle)\langle\psi_{j+1}|$. We set $N(\omega_j) = 1/(e^{\omega_j/(k_B T)} - 1)$ with $\omega_j = E_j - E_{j-1}$ so that the system relaxes to the equilibrium state of $\mathcal{H}_0$ at temperature T.

A many-spin model with a spin-nematic phase is surely superior to the nanospin model (1) for the purpose of studying magnon-pair resonance. However, there are at least three reasons why the model (1) is expected to capture the essential aspect of a magnon-pair resonance in bulk systems. First, the diffraction limit (∼wavelength) of a THz laser is much larger than the lattice space of the magnets and therefore only magnetic excitations around the wave number $\boldsymbol{k} = \boldsymbol{0}$ are relevant for a laser application. Even bulk magnets have only a few discrete modes around $\boldsymbol{k} = \boldsymbol{0}$ [22–25] and excited states in the model (1) may be viewed as analogs of these $\boldsymbol{k} = \boldsymbol{0}$ modes. Second, the positions of two magnons in a single magnon pair are quite close to each other [44,45] because the attractive force between two magnons stems from short-range exchanges. Therefore, excited states with two (one) down-spins in the model (1) are analogous to those with a magnon pair (magnon) in a bulk magnet. The third point, which is most important, is that one can practically take the dissipation effect into account within the Lindblad approximation if the spin system is small enough. For the analysis of realistic magnetic-resonance spectra, small interactions breaking spin conservation and the spin-bath coupling (e.g., magnetic anisotropies, dipole interactions, spin-phonon coupling, etc.) are more important than many-body effects. In fact, the observed ESR spectrum shapes of nanomagnets [67–69] are often similar to those of bulk magnets [22–25]. The Lindblad term phenomenologically describes the effect of such small interactions and makes the system relax to the equilibrium state. For correlated many-spin systems, even finding energy eigenstates is difficult and treating the dissipation effect in such bulk systems is a massively hard task. Moreover, if we continuously apply a laser to an isolated many-spin system decoupled to the environment, the system is generally heated up. From these arguments, we discuss magnon-pair resonance by using the model (1) with the quantum master equation (2). The analysis of spin dynamics in dissipative *many-spin* systems is left to a future study.

We note that the magnon-pair band is sometimes located around a wave number $k^\alpha = \pi$ ($\alpha = x$, y, or z) in *antiferromagnetic* spin-nematic magnets [44–47,49,50]. Magnon pairs on such a band do not seem to be coupled to applied THz or GHz electromagnetic waves. However, even in those cases, if the crystal symmetry is low enough and the unit cell includes multiple magnetic ions (e.g., due to dimerization), the usual ac Zeeman coupling of a THz laser can excite magnon pairs. In addition, if magnetoelectric coupling [70] exists in the spin-nematic magnets, excitations around $k^\alpha = \pi$ can be often created with a laser [11–13,28,29].

Analysis and results. Based on the master Eq. (2), we study laser-driven magnetic resonance in the model (1) with $J_1 = -2.2$, $J_2 = 1$, $\Delta_1 = \Delta_2 = 0.24$, and $H = 0.56$. We consider the low-temperature range of $k_B T \lesssim 0.1$. The initial state at $t = 0$ is set to be a polarized equilibrium state with a temperature T and then we add the ac Zeeman coupling $\mathcal{H}_{\rm cp}$ or $\mathcal{H}_{\rm lp}$. The Lindblad term helps the laser-driven system to return to the equilibrium state at T. From the low-energy levels of Eq. (1) shown in Fig. 2, one sees that $|\psi_1\rangle$, $|\psi_2\rangle$, and $|\psi_3\rangle$ have $S^z_{\rm tot} = 2$, 1, and 0, respectively. Therefore, we may view $|\psi_2\rangle$ and $|\psi_3\rangle$ as magnon and magnon-pair states, respectively. As we mentioned, magnons (magnon pairs) can be resonantly

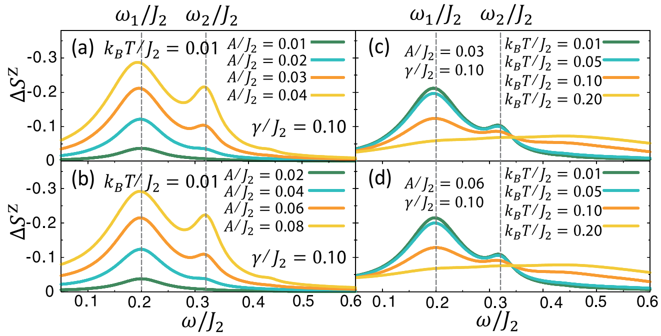

FIG. 3. Spectra $\Delta S^z(\omega)$ for different values of laser strength or temperature in the case of (a), (c) a circularly polarized laser and (b), (d) a linearly polarized laser. Parameters are set to be $J_1 = -2.2$, $J_2 = 1$, $\Delta_1 = \Delta_2 = 0.24$, and $H = 0.56$. Dotted lines denote the resonant positions at $\omega = \omega_1$ and ω_2.

excited by a single photon (two photons). Thus the frequency of the magnon resonance is given by $\omega_1 = E_2 - E_1 = H - 1.5\Delta_1 = 0.2$, while that of magnon-pair resonance is $\omega_2 = (E_3 - E_1)/2 = H - \Delta_1 = 0.32$, as shown in Fig. 2(b). We will consider the range of laser frequency ω including these resonant values ω_1 and ω_2. We note that $\omega_2 - \omega_1 = 0.12$ is much smaller than $J_2 = 1$ and it means that our setup imposes a tough condition to distinguish two resonance peaks.

One can numerically calculate the expectation value of any operator $\mathcal{O}$ at arbitrary time t from the density matrix $\rho(t)$: $\langle \mathcal{O}(t)\rangle = {\rm Tr}[\mathcal{O}\rho(t)]$. Here, we concentrate on the magnetization change between the initial state and the nonequilibrium steady one [66] which is realized by waiting for a long time from the beginning of the laser application. Namely, we compute $\Delta S^z = \langle S^z_{\rm tot}\rangle_{\rm neq} - \langle S^z_{\rm tot}\rangle_{\rm eq}$, where

$$\langle S^z_{\rm tot}\rangle_{\rm neq} = \frac{1}{\tau_0}\int_\tau^{\tau+\tau_0} dt \langle S^z_{\rm tot}(t)\rangle, \tag{3}$$

and $\langle S^z_{\rm tot}\rangle_{\rm eq}$ is the initial value of $\langle S^z_{\rm tot}\rangle$ at a fixed $k_B T$. Here, τ and τ_0 are set to be sufficiently larger than the relaxation time $\hbar/\gamma$ and the period $2\pi/\omega$ of the laser, respectively. The integration of Eq. (3) is necessary to eliminate the small fluctuation of $\langle S^z_{\rm tot}(t)\rangle$, especially in the case of a linearly polarized laser. A large $|\Delta S^z|$ indicates a large precession motion (i.e., a large oscillation of transverse magnetization) driven by the laser [15] and it means that the system efficiently absorbs photons. Therefore, we can use $|\Delta S^z|$ as an index of the observability of magnon-pair resonances.

The relaxation time of electron spins in solids is usually from picoseconds to nanoseconds [6,19–21,71–76]. The current THz-laser technique enables us to utilize intense THz-laser pulses with a few T, which corresponds to a few MV/cm [1–4]. For the reality, we hence consider the range of $\gamma \sim 0.01J_2$–$0.1J_2$ and $A/J_2 \lesssim 0.1$ in the numerical calculation of ΔS^z: For instance, for $J_2/k_B = 10$ K (50 K), $\gamma = 0.05J_2$ and $A = 0.05J_2$ respectively correspond to the relaxation time $\hbar/\gamma \simeq 15.2$ ps (3.0 ps) and the ac magnetic field $h_{\rm ac} \simeq 0.37$ (1.86) T.

Figure 3 depicts the computed $\Delta S^z(\omega)$ as a function of ω, changing the laser strength A or temperature T. We find that

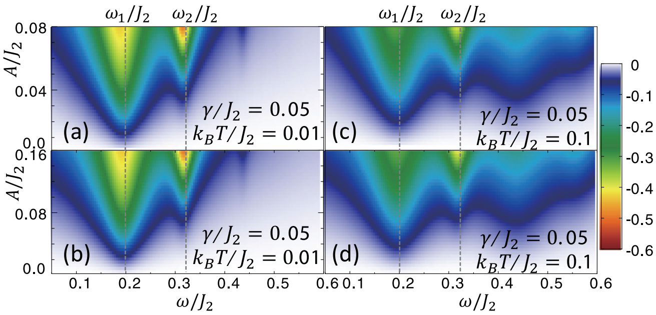

FIG. 4. Laser-strength A and frequency ω dependences of the spectra $\Delta S^z(\omega)$ in the case of (a), (c) a circularly polarized laser and (b), (d) a linearly polarized one. We set $J_1 = -2.2$, $J_2 = 1$, $\Delta_1 = \Delta_2 = 0.24$, and $H = 0.56$. Dotted lines denote the resonant positions at $\omega = \omega_1$, and ω_2.

when the laser is weak in low T, only the magnon resonance at $\omega = \omega_1$ is clearly observed (this corresponds to the standard magnetic resonance), while magnon-pair peaks gradually grow up with an increase of A. One sees that magnon-pair peaks become visible for $A \gtrsim 0.03J_2$ ($A \gtrsim 0.06J_2$) in the case of a circularly (linearly) polarized laser. Therefore the result of Fig. 3 indicates that the magnon-pair resonance can be observed with an available strong laser or electromagnetic wave. Figure 4 shows the laser-induced magnetization ΔS^z for both circularly and linearly polarized lasers in a large range of (A, ω). This figure also tells us that magnon-pair peaks at $\omega = \omega_2$ become visible if the applied wave is strong enough.

To more quantitatively see the required intensity of the laser, we show the laser-strength dependence of ΔS^z at the resonant points $\omega = \omega_1$ and ω_2 in Fig. 5. We find that the magnon-resonance peak almost linearly increases with A, especially in a weak dissipation regime, whereas the magnon-pair peak exhibits a nonlinear increase in terms of A. Moreover, the magnon-pair peak becomes comparable to the magnon one if A is sufficiently strong ($A \gtrsim 0.01J_2 - 0.05J_2$).

Finally, we introduce another index for the visibility of magnon-pair resonances. As shown in Fig. 6(a), we first

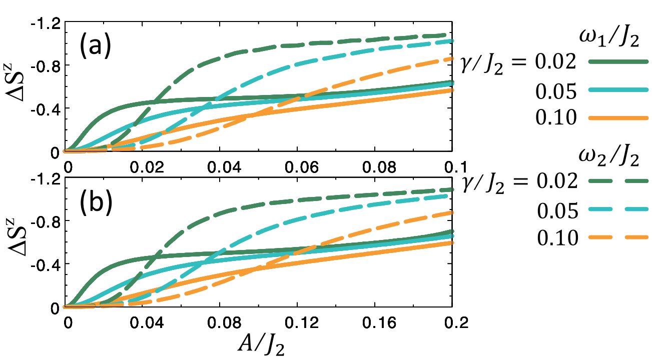

FIG. 5. Laser strength dependence of the difference $\Delta S^z(\omega)$ at the resonant points $\omega = \omega_1$ and ω_2 for (a) a circularly and (b) linearly polarized laser. We use the parameters $J_1 = -2.2$, $J_2 = 1$, $\Delta_1 = \Delta_2 = 0.24$, $H = 0.56$, and $k_BT = 0.01$.

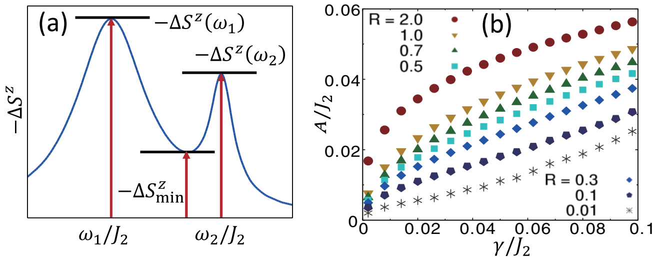

FIG. 6. (a) Definition of $\Delta S^z_{\rm min}$. (b) Contour line of the ratio R of Eq. (4) in $(\gamma/J_2, A/J_2)$ space in the case of a circularly polarized laser. We again set $J_1 = -2.2$, $J_2 = 1$, $\Delta_1 = \Delta_2 = 0.24$, $H = 0.56$, and $k_BT = 0.01$. We also obtain a similar contour line in the case of a linearly polarized laser.

define $-\Delta S^z_{\rm min}$ as the minimum value of $|\Delta S^z(\omega)|$ in the range between $\omega = \omega_1$ and ω_2. Using it, let us consider the following quantity,

$$R(\omega_1, \omega_2) = \frac{\Delta S^z(\omega_2) - \Delta S^z_{\rm min}}{\Delta S^z(\omega_1) - \Delta S^z_{\rm min}}. \tag{4}$$

A sufficient large R means a high possibility of detecting a magnon-pair peak in the real experiment. Figure 6(b) draws the contour curve of R in (γ, A) space, and it clearly indicates that a large laser strength A and a small dissipation constant γ are better for the observation of magnon-pair resonance. If we assume that $R > 0.1$ is the necessary condition for the observation of a magnon-pair peak, Fig. 6(b) implies that the peak can be observed in the region of $A \gtrsim 0.3\gamma$.

From Figs. 3–6, we conclude that one can observe not only magnon but also magnon-pair resonances in fully polarized states of spin-nematic magnets if the laser intensity reaches $h_{\rm ac} \sim 0.1$–1.0 T and the resonant points $\omega_{1,2}$ are sufficiently separated.

Conclusions. In summary, we theoretically discussed the observability of magnon-pair resonance in fully polarized states of spin-nematic magnets. We compute the laser-driven spin dynamics in the frustrated nanospin model of Eq. (1), which is analogous to a bulk spin-nematic magnet, by applying the Lindblad equation. Our calculation strongly indicates that a currently available intense laser with $h_{\rm ac} \lesssim 1$ T is enough to observe magnon-pair resonances. Besides spin-nematic or nanomagnets, bound states of magnons also emerge in a class of frustrated or low-dimensional quantum magnets [77,78]. Our estimation of the required laser strength for magnon-pair resonances would be applicable to such magnets.

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