

Spin-caloritronic signatures of soft magnons in bilayer CrSBr

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Spin transport in magnetic insulators is often treated by assuming that magnons carry a fixed spin angular momentum of $\hbar$, which does not hold in general, however. Here, we calculate the magnon spin angular momentum of a layered antiferromagnet as a function of applied magnetic field and wave vector. We show that the triaxial anisotropy and intralayer dipolar interactions in bilayer CrSBr renormalize the magnon spin angular momentum, which diverges upon field-induced magnon softening. This divergence gives rise to a pronounced peak in the thermal spin Seebeck response and provides a clear spin-caloritronic signature of soft magnons.

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I. INTRODUCTION

The spin current generated by thermal gradients in magnetic insulators is a central theme in spin caloritronics. When injected into a metal contact, a transverse voltage builds up due to the inverse spin Hall effect [1–4], as observed in a wide range of mainly oxidic magnetic insulators [5–8].

Within this broader class of magnetic insulators, single or multilayered two-dimensional (2D) van der Waals (vdW) magnets offer an attractive platform for spintronics. The dependence of spin transport on external magnetic and electric fields and stacking geometry [9–11] may provide valuable information on the carriers of spin and energy, *viz.* the quanta of the spin waves, or magnons. The relatively weak interlayer coupling in vdW multilayers causes magnetic properties to differ strongly from those of bulk materials [12,13]. The gate-tunable interlayer exchange demonstrated in CrI₃ [14], for example, highlights the potential of vdW magnets for energy-efficient spintronic devices.

Among these vdW magnets, CrSBr is a particularly interesting magnetic insulator with a tetragonal crystal lattice structure. It displays intralayer ferromagnetic (FM) and interlayer antiferromagnetic (AFM) coupling [15] and strong triaxial magnetocrystalline anisotropy [16–18]. Its bilayer magnetic textures and excitations have been accurately described in Refs. [19,20]. The presence of (A)FM resonance frequencies in the GHz range [19,20], together with its air-stable nature [17], makes it a promising candidate for spintronic applications. Despite a growing body of work on magnetic order and excitations, the implications of the magnon mode structure for spin transport in CrSBr remain largely unexplored.

On a microscopic level, the spin-caloritronic response depends on the magnon band structure and the mode-dependent spin angular momentum (SAM) [21]. The magnon SAM is often assumed to be $\hbar$ [22–24] but this need not be the case in the presence of magnetic anisotropy and dipole-dipole interactions [21,25,26], both of which play an important role in CrSBr [19]. This deviation of the SAM from $\hbar$ arises because anisotropy and dipolar interactions mix magnon creation and annihilation operators, so a quasiparticle is no longer a circularly polarized spin-lowering excitation carrying exactly $\hbar$. The SAM becomes mode- and wave-vector-dependent and can be strongly enhanced when the mode softens [21]. Moreover, CrSBr exhibits field-induced magnon softening at experimentally accessible magnetic fields [17], making it a particularly suitable platform for studying the consequences of renormalized magnon SAM for spin-caloritronic transport.

The central question of this work is how the magnon SAM deviates from the commonly assumed value of $\hbar$ in mono- and bilayers of CrSBr and how this nonuniversality modifies the thermal and diffusive spin Seebeck coefficients (SSCs). The thermal SSC describes magnon spin transport driven by a temperature gradient, whereas the diffusive SSC describes transport driven by a magnon chemical potential gradient associated with a spatially nonuniform magnon accumulation. To investigate these responses, we first determine the magnon frequencies and eigenvectors from the quadratic spin-wave Hamiltonian, and calculate the mode-resolved SAM from these eigenvectors. We then incorporate the resulting magnon dispersion and SAM into a Boltzmann description of thermal and diffusive spin transport. The monolayer serves as a reference system that isolates the soft-mode mechanism, while the bilayer additionally captures the relative spin contributions from the two magnetic layers.

The paper is organized as follows. In Sec. II, we start from the Hamiltonians introduced in Ref. [19] for mono- and bilayer CrSBr. In Sec. III, we calculate the magnon SAM as the expectation value of the spin operator for magnon eigenstates. In Sec. IV, we compute the SSC under in-plane external fields and gradients of temperature and chemical potential. Section V contains a summary and discussion.

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II. THEORETICAL FRAMEWORK

Throughout this work, we adopt the model and material parameters from Ref. [19], where we already computed the magnon spectrum. To compute the magnon spin, we require, here, also the magnon wave functions. Figure 7 shows the top view of a CrSBr monolayer. The Hamiltonian for the monolayer (bilayer) is shown in Appendix B (C). As in our previous work [19], we model each monolayer as an in-plane ferromagnet with a single spin per unit cell. This approximation is justified because the high-energy band associated with out-of-phase intralayer excitations (see Appendixes A and B of Ref. [19]) contributes negligibly to the spin transport. Here, we focus on the magnetodipolar terms. For a thin film with in-plane magnetization oriented along the $\hat{e}_\gamma$ axis, where γ is a Cartesian coordinate and i ($= A, B$) the layer index (see Appendixes B 3 and C 3), the dynamic dipolar field at site j is given by [27]

$$\begin{aligned} \vec{B}_{\text{dip},j,i}(\vec{k}) = & -\mu_0 M_s f(k) m_{\beta,j,i} \hat{e}_\beta^{(i)} \\ & -\mu_0 M_s \frac{k_{\alpha,i}^2}{k^2} [1 - f(k)] m_{\alpha,j,i} \hat{e}_\alpha^{(i)}, \end{aligned} \quad (1)$$

where μ_0 is the vacuum permeability, M_s is the saturation magnetization, the $\alpha(\beta)$ direction is the in-plane (out-of-plane) Cartesian component perpendicular to γ , and $f(k) = (1 - e^{-kd})/(kd)$, where $k = |\vec{k}|$ and the thickness of the monolayer $d \approx b/2$ [19] with b the easy axis lattice vector. The magnetic moment $\vec{m}_j = [m_{\alpha,j}, m_{\beta,j}, m_{\gamma,j}]^T = \gamma \hbar \vec{S}_j / M_s$, where γ is the gyromagnetic ratio (not to be confused with subscript γ for the Cartesian coordinate). The Zeeman interaction of the magnetic moments with the dipolar field can be written as a spin Hamiltonian [see Eqs. (B1) and (C1)],

$$\begin{aligned} H_{\text{dip},i}(\vec{k}) = & \sum_j \left(\frac{1}{2} \mu_0 \frac{\gamma \hbar M_s}{S} f(k) (S_{j,i}^{(\beta)})^2 \right. \\ & \left. + \frac{1}{2} \mu_0 \frac{\gamma \hbar M_s}{S} \frac{k_{\alpha,i}^2}{k^2} (1 - f(k)) (S_{j,i}^{(\alpha)})^2 \right). \end{aligned} \quad (2)$$

We showed previously that the intralayer dipolar and crystal anisotropy fields are of the same order of magnitude [19]. Corrections due to the discrete lattice can be incorporated through an Ewald sum of the dipole-dipole interactions [28], but are expected to be small and are, therefore, neglected here.

A. Monolayer

The monolayer magnon Hamiltonian can be derived from the spin Hamiltonian in Appendix B 1 via the Holstein-Primakoff (HP) transformation [29]

$$\begin{aligned} S_j^{(\gamma)} &= S - \hat{a}_j^\dagger \hat{a}_j, \\ S_j^{(\alpha)} &= \sqrt{\frac{S}{2}} (\hat{a}_j + \hat{a}_j^\dagger), \\ S_j^{(\beta)} &= -i\sqrt{\frac{S}{2}} (\hat{a}_j - \hat{a}_j^\dagger), \end{aligned} \quad (3)$$

where α and β are the transverse components to γ . To lowest order in the magnon number with $\hat{a}_i = \frac{1}{\sqrt{N}} \sum_k \hat{a}_k e^{i\vec{k} \cdot \vec{r}_i}$ the

TABLE I. Magnetic phases and corresponding indices l for monolayer CrSBr.

Phase	Index l
FM	m_{FM}
Canted	m_{cant}
Saturated	m_{sat}

Hamiltonian reads

$$H^{(l)} = - \sum_k \left[A_k^{(l)} \hat{a}_k^\dagger \hat{a}_k + \frac{B_k^{(l)}}{2} (\hat{a}_k \hat{a}_{-k} + \hat{a}_k^\dagger \hat{a}_{-k}^\dagger) + C_k^{(l)} \right], \quad (4)$$

where l indicates the magnetic phase in the monolayer (see Table I).

The functions A_k and B_k are shown in Appendix B 4 and $\sum_k C_k^{(l)}$ contributes a constant shift that can be disregarded. Because Eq. (4) contains anomalous pair-creation and pair-annihilation terms, it is not diagonal in the HP magnon basis. We, therefore, introduce the Bogoliubov transformation for the monolayer [30] $\Phi_1 = T_1 \Psi_1$, where

$$\Psi_1 = \begin{pmatrix} \hat{a}_k \\ \hat{a}_{-k}^\dagger \end{pmatrix}, \quad \Phi_1 = \begin{pmatrix} \hat{\alpha}_k \\ \hat{\alpha}_{-k}^\dagger \end{pmatrix}, \quad T_1 = \begin{bmatrix} u_k & v_k \\ v_k & u_k \end{bmatrix}. \quad (5)$$

Equation (4) reads

$$H^{(l)} = \frac{1}{2} \Psi_1^\dagger \mathcal{H}^{(l)} \Psi_1 + \text{constant}, \quad (6)$$

where

$$\mathcal{H}^{(l)} = \begin{bmatrix} -A_k^{(l)} & -B_k^{(l)} \\ -B_k^{(l)} & -A_k^{(l)} \end{bmatrix} \quad (7)$$

and the factor $\frac{1}{2}$ corrects for double counting. The elements of both the original ($\hat{a}$) and quasiparticle ($\hat{\alpha}$) operator vectors obey the bosonic commutation relations $[\Psi_{1,i}, \Psi_{1,j}^\dagger] = [\Phi_{1,i}, \Phi_{1,j}^\dagger] = \Sigma_{1,ij}$ [31], where

$$\Sigma_1 = \begin{bmatrix} 1 & 0 \\ 0 & -1 \end{bmatrix}. \quad (8)$$

The eigenvalues of $\Sigma_1 \mathcal{H}^{(l)}$ give the magnon spectrum $\omega_\alpha^{(l)} = \sqrt{(A_k^{(l)})^2 - (B_k^{(l)})^2}$ [32], while

$$T_1 \Sigma_1 \mathcal{H}^{(l)} T_1^{-1} = \Sigma_1 \mathcal{E}^{(l)}. \quad (9)$$

with $\mathcal{E}^{(l)} = \text{diag}(\omega_\alpha^{(l)}, \omega_\alpha^{(l)})$. Moreover, the j th column of T_1^{-1} is an eigenvector of $\Sigma_1 \mathcal{H}^{(l)}$ so that

$$\Sigma_1 \mathcal{H}^{(l)} \begin{pmatrix} u_k \\ -v_k \end{pmatrix} = \omega_\alpha^{(l)} \begin{pmatrix} u_k \\ -v_k \end{pmatrix}. \quad (10)$$

In summary, the monolayer provides the reference system in which anisotropy and dipolar interactions mix magnon creation and annihilation operators through the Bogoliubov transformation. The resulting eigenfrequencies determine the magnon dispersion, and the coefficients u_k and v_k quantify the polarization of each mode. These quantities form the microscopic input for calculating the nonuniversal magnon SAM in Sec. III and the resulting SSC in Sec. IV.

B. Bilayer

In the bilayer, each spin is transformed in its respective static magnetization frame via the HP transformations:

$$\begin{aligned} S_j^{(\gamma,A)} &= S - \hat{a}_j^\dagger \hat{a}_j, S_j^{(\gamma,B)} = S - \hat{b}_j^\dagger \hat{b}_j, \\ S_j^{(\alpha,A)} &= \sqrt{\frac{S}{2}}(\hat{a}_j + \hat{a}_j^\dagger), S_j^{(\alpha,B)} = \sqrt{\frac{S}{2}}(\hat{b}_j + \hat{b}_j^\dagger), \\ S_j^{(\beta,A)} &= -i\sqrt{\frac{S}{2}}(\hat{a}_j - \hat{a}_j^\dagger), S_j^{(\beta,B)} = -i\sqrt{\frac{S}{2}}(\hat{b}_j - \hat{b}_j^\dagger), \end{aligned} \quad (11)$$

where $\hat{a}_j^{(\dagger)}$ and $\hat{b}_j^{(\dagger)}$ are the magnon operators for layers A and B, respectively. In the bilayer, the interlayer exchange additionally hybridizes the spin-wave excitations of the two layers. The resulting modes are, therefore, superpositions of particle and hole amplitudes in both layers, whose relative weights determine the contributions to their SAM. The Bogoliubov transformation for the bilayer is $\Phi_2 = T_2 \Psi_2$, where

$$\begin{aligned} \Psi_2 &= \begin{pmatrix} \hat{a}_k \\ \hat{b}_k \\ \hat{a}_{-k}^\dagger \\ \hat{b}_{-k}^\dagger \end{pmatrix}, \quad \Phi_2 = \begin{pmatrix} \hat{\alpha}_k \\ \hat{\beta}_k \\ \hat{\alpha}_{-k}^\dagger \\ \hat{\beta}_{-k}^\dagger \end{pmatrix}, \\ T_2 &= \begin{bmatrix} u_a^{(\alpha)} & u_b^{(\alpha)} & v_a^{(\alpha)} & v_b^{(\alpha)} \\ u_a^{(\beta)} & u_b^{(\beta)} & v_a^{(\beta)} & v_b^{(\beta)} \\ v_a^{(\alpha)} & v_b^{(\alpha)} & u_a^{(\alpha)} & u_b^{(\alpha)} \\ v_a^{(\beta)} & v_b^{(\beta)} & u_a^{(\beta)} & u_b^{(\beta)} \end{bmatrix}, \end{aligned} \quad (12)$$

with $[\Psi_{2,i}, \Psi_{2,j}^\dagger] = [\Phi_{2,i}, \Phi_{2,j}^\dagger] = \Sigma_{2,ij}$ and

$$\Sigma_2 = \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & -1 & 0 \\ 0 & 0 & 0 & -1 \end{bmatrix}. \quad (13)$$

This leads to an equation analogous to Eq. (9) with eigenvalues in $\mathcal{E}^{(l)} = \text{diag}(\omega_a^{(l)}, \omega_b^{(l)}, \omega_a^{(l)}, \omega_b^{(l)})$ that solve an equation similar to Eq. (10):

$$\Sigma_2 \mathcal{H}^{(l)} \begin{pmatrix} u_a^m \\ u_b^m \\ -v_a^m \\ -v_b^m \end{pmatrix} = \omega_m^{(l)} \begin{pmatrix} u_a^m \\ u_b^m \\ -v_a^m \\ -v_b^m \end{pmatrix}, \quad (14)$$

where $m = \alpha, \beta$ corresponding to the two magnon bands of the bilayer. The bilayer formalism extends the monolayer treatment to two coupled magnetic layers, resulting in two magnon branches. The corresponding Bogoliubov eigenvectors describe the magnon amplitudes in the two layers for each magnetic phase. The SAM of each mode is obtained by combining the contributions from layers A and B according to their relative spin alignment. We calculate the resulting field-dependent SAM in Sec. III B. Table II shows the magnetic phases and corresponding indices l in the bilayer. We show $\mathcal{H}^{(l)}$ for all phases in the bilayer in Appendix C 4.

TABLE II. Magnetic phases and corresponding indices l for bilayer CrSBr.

Phase	Index l
AFM	b_{AFM}
FM	b_{FM}
Canted	b_{cant}
Saturated	b_{sat}

III. SPIN ANGULAR MOMENTUM

In this section, we evaluate the nonuniversal magnon SAM in both the mono- and bilayer by computing the expectation value of the static magnetization in the diagonal Bogoliubov basis. We first consider the monolayer in order to isolate the effects of intralayer anisotropy and dipolar interactions, without the additional mode hybridization in the bilayer.

A. Monolayer

Since a Bogoliubov quasiparticle is a superposition of magnon creation and annihilation operators, its SAM is not quantized to $\hbar$. We, therefore, define its SAM as the change in the spin expectation value upon creating a quasiparticle relative to the Bogoliubov vacuum. The SAM carried by a single magnon in the monolayer branch α , with the spin quantization axis γ along the equilibrium magnetization, is [26]

$$\Delta S_\alpha^{(\gamma)} = \langle 1_\alpha | S^{(\gamma)} | 1_\alpha \rangle - \langle 0 | S^{(\gamma)} | 0 \rangle, \quad (15)$$

where $|0\rangle$ is the vacuum with $\hat{\alpha}_k |0\rangle = 0$ and $|1_\alpha\rangle = \alpha_k^\dagger |0\rangle$. Combining Eqs. (3), (5), and Eq. (15) leads to $\Delta S_\alpha^{(\gamma)} = -|u|^2$ in units of $\hbar$. Combining Eq. (10) together with the bosonic commutation condition $|u|^2 - |v|^2 = 1$ ($\hat{e}_j^T \Sigma_1 \hat{e}_j = 1$) we find

$$\Delta S_\alpha^{(\gamma,l)} = -|u^{(l)}|^2 = -\frac{1}{1 - \left(\frac{\omega_a^{(l)} + A_k^{(l)}}{B_k^{(l)}} \right)^2}. \quad (16)$$

In the canted configuration (see Appendix B 3), the SAM along the x and z axes are $-|u^{(l)}|^2 \sin \theta$ and $-|u^{(l)}|^2 \cos \theta$, respectively.

An external field along the easy axis (b) of the monolayer leads to the collinear configuration ($\gamma = z, l = m_{\text{FM}}$) in which the magnetization direction is along z . A field along the intermediate axis (a) with $B_0 < B_{\text{sat}}$ cants the spins at an angle between the b and a axes ($l = m_{\text{cant}}$), while for $B_0 \geq B_{\text{sat}}$ the magnetization is along the a axis ($\gamma = x, l = m_{\text{sat}}$). Appendix B 4 shows the matrix elements of $\mathcal{H}^{(l)}$ and Fig. 1 the field dependence of the SAM for the Kittel mode at $k = 0$ since softening occurs at the Brillouin-zone center and low-wave-vector magnons dominate the low-temperature response.

The red curve corresponds to collinear spin and external field along the easy axis (b). Here, a SAM larger than $\hbar$ indicates the degree of elliptical polarization [33] by the triaxial anisotropy and intralayer dipole interactions [19]. In the limit of a large applied magnetic field $|\Delta S^{(\gamma)}| \rightarrow \hbar$.

The blue curves are the results for an external field along the intermediate axis (a). The solid blue curve is the projection of the SAM along a , and the dotted curve along b ,

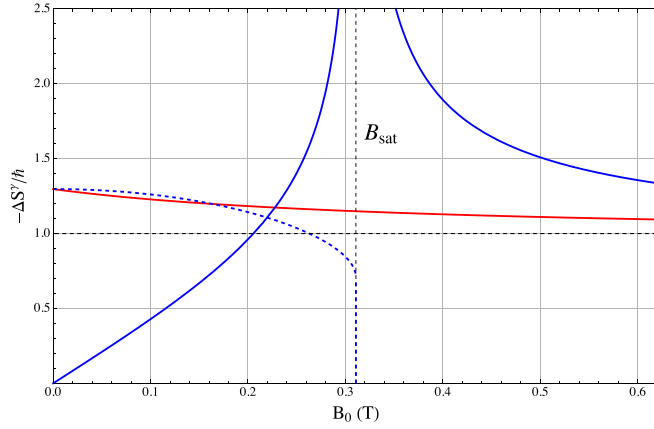


FIG. 1. Calculated SAM of the Kittel mode ($k = 0$) in the monolayer as a function of the external magnetic field B_0 . The red curve shows the SAM along the easy axis ($\vec{B}_0 \parallel \hat{b}$) while the blue ones hold for $\vec{B}_0 \parallel \hat{a}$ in the canted ($B_0 < B_{\text{sat}}$) and saturated ($B_0 \geq B_{\text{sat}}$) phases: the solid (dotted) line shows the projection of the SAM along the a axis (b axis).

perpendicular to the external field. For fields approaching the saturation value the SAM increases strongly and ultimately diverges when the magnon frequency softens to zero, reflecting an increasingly elliptical precession [21]. This ellipticity arises from the different energy scales associated with the two directions perpendicular to the equilibrium magnetization, resulting in unequal amplitudes of the transverse magnetization components. Microscopically, this asymmetry gives rise to anomalous (pair-creation and pair-annihilation) terms in the bosonic Hamiltonian, which mix magnon creation and annihilation operators through the Bogoliubov transformation. Consequently, the magnon eigenstate is no longer a pure spin-lowering excitation, and its spin expectation value deviates from $\hbar$. Since the intralayer dipolar field effectively renormalizes the anisotropy constants, either triaxial magnetic anisotropy or intralayer dipole-dipole interactions alone can produce this imbalance of transverse energy scales and thereby induce elliptical precession and, consequently, a divergent SAM. At the critical soft point ($\omega \rightarrow 0$) the magnon becomes linearly polarized. Here, the precession amplitudes diverge because the anisotropy and magnetic-field torques cancel. This divergence of the *spin-wave amplitude* is unphysical and signals the breakdown of linear spin-wave theory: close to B_{sat} the small-amplitude assumption underlying the lowest-order HP expansion is no longer valid, and higher-order terms describing magnon-magnon interactions must be taken into account. While these interactions are expected to regularize the divergent spin-wave amplitude, the enhancement of the magnon SAM and the corresponding peak in the SSC are expected to remain robust. Far above B_{sat} , the SAM approaches $\hbar$.

The monolayer results establish the microscopic origin of the nonuniversal magnon SAM. For an easy-axis field, the magnon remains relatively hard and its SAM approaches $\hbar$ as the field increases. For an intermediate-axis field, however, the mode softens at B_{sat} , producing strongly elliptical polarization and a pronounced enhancement of the SAM within linear spin-wave theory. This field-controlled enhancement provides

the mechanism for the spin-caloritronic signature calculated in Sec. IV A.

B. Bilayer

In the bilayer, the SAM of each mode combines the layer-resolved spin contributions, which add for parallel alignment and subtract for antiparallel alignment. Thus, for parallel (+) or antiparallel (−) spin alignment, the SAM of mode m ($= \alpha, \beta$) reads

$$\Delta S_m^{(\gamma)} = \langle 1_m | S^{(\gamma,A)} \pm S^{(\gamma,B)} | 1_m \rangle - \langle 0 | S^{(\gamma,A)} \pm S^{(\gamma,B)} | 0 \rangle, \quad (17)$$

where in the vacuum $\hat{m}_k |0\rangle = 0$ and $|1_m\rangle = \hat{m}_k^\dagger |0\rangle$, with $\hat{m}_k^{(\dagger)}$ the quasi-particle operator from Eq. (12). From Eqs. (11), (12) and (17) it follows that $\Delta S_\alpha^{(\gamma,l)} = -(|u_a^{(\alpha,l)}|^2 \pm |u_b^{(\alpha,l)}|^2)$ and $\Delta S_\beta^{(\gamma,l)} = -(|u_a^{(\beta,l)}|^2 \pm |u_b^{(\beta,l)}|^2)$ in units of $\hbar$, for which we combine Eq. (14) with the bosonic commutation relation $|u|^2 - |v|^2 = 1$ ($\hat{e}_j^T \Sigma \hat{e}_j = 1$).

In the canted configuration (see Appendix B3) the z components cancel by symmetry so that the effective SAM along the x axis is $-\sin \theta (|u_a^{(\alpha,l)}|^2 + |u_b^{(\alpha,l)}|^2)$ and $-\sin \theta (|u_a^{(\beta,l)}|^2 + |u_b^{(\beta,l)}|^2)$ for the α and β modes, respectively.

In the bilayer we distinguish four phases depending on the direction and magnitude of the applied field. When a field along the easy axis (b) stays below the spin-flip transition ($B_0 < B_{\text{crit}}^{\text{flip}}$), the AFM phase ($\gamma = \pm z, l = b_{\text{AFM}}$) is stable. Here, the spin are aligned anti-parallel along the easy axis. When $B_0 \geq B_{\text{crit}}^{\text{flip}}$ the magnetization of both layers are parallel along the external field ($\gamma = z, l = b_{\text{FM}}$). Appendix C4 derives $\mathcal{H}^{(l)}$ in Eq. (14) for the AFM and FM phases in the bilayer, while Fig. 2(a) shows the corresponding SAM as a function of the external magnetic field applied along the easy axis. In the AFM regime, the two magnon branches carry opposite SAM [34]. For small external fields the SAM is close to zero due to canceling z components of comparable magnitude of the magnetization. At $B_0 = B_{\text{crit}}^{\text{flip}}$, the system undergoes a spin-flip transition to the FM phase. In both phases the anisotropies cause deviations from $\hbar$ as in the monolayer for $\vec{B}_0 \parallel \hat{b}$.

Applying a field along the intermediate axis (a) symmetrically cants the spins from the $\pm b$ axes to the a axis ($B_0 < B_{\text{sat}}^{\text{cant}}, l = b_{\text{cant}}$), see Appendix C3. At the saturation field $B_0 \geq B_{\text{sat}}^{\text{cant}}$ all spins align with the a axis ($\gamma = x, l = b_{\text{sat}}$). Appendix C4 derives $\mathcal{H}^{(l)}$ in Eq. (14) for the canted and saturated phases in the bilayer, and Fig. 2(b) shows the SAMs projected along the intermediate axis as a function of the external magnetic field. In the canted phase an increasing external field leads to the blue branch softening with enhanced ellipticity and SAM, analogous to the magnon mode in the monolayer. The algebraic divergence is again not physical but indicates the breakdown of the HP expansion. The second (orange curve) branch remains at high-frequencies due to interlayer exchange with relatively small effects of the anisotropies for all field values, resulting in a weakly field-dependent SAM. At $B_0 = B_{\text{sat}}^{\text{cant}}$, the system undergoes a transition to the saturated phase, where all spins become collinear with the external

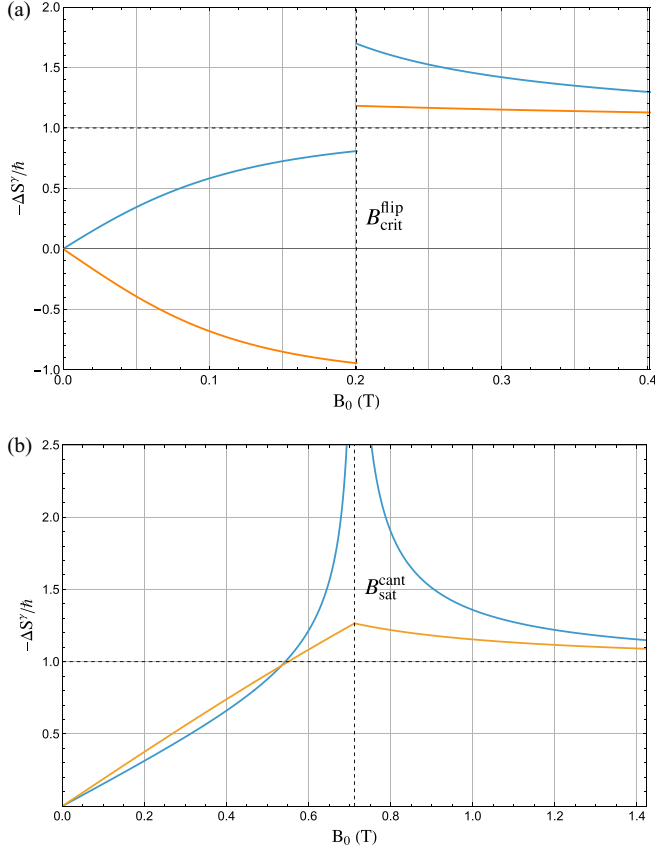


FIG. 2. SAM of the magnon modes at $k = 0$ in the bilayer as a function of the external magnetic field B_0 . The lower (higher) frequency band is indicated in blue (orange). (a) SAM along the easy axis (b) in the AFM ($B_0 < B_{\text{crit}}^{\text{flip}}$) and FM ($B_0 \geq B_{\text{crit}}^{\text{flip}}$) phases ($\vec{B}_0 \parallel \hat{b}$). (b) SAM along the intermediate axis (a) in the canted ($B_0 < B_{\text{sat}}^{\text{cant}}$) and saturated ($B_0 \geq B_{\text{sat}}^{\text{cant}}$) phases ($\vec{B}_0 \parallel \hat{a}$).

field. Beyond this point, the magnon spectrum hardens and the SAM of both modes gradually approaches $\hbar$.

The bilayer combines the soft-mode mechanism found in the monolayer with additional effects due to the relative spin contributions. For an easy-axis field, the two branches carry opposite and nearly compensating SAM in the AFM phase, whereas the spin-flip transition aligns their spin contributions in the FM phase. For an intermediate-axis field, the lower branch softens and acquires a strongly enhanced SAM near $B_{\text{sat}}^{\text{cant}}$, while the upper branch remains comparatively hard. These contrasting mode characteristics determine which contributions cancel or dominate the spin Seebeck response discussed in Sec. IV B.

IV. SPIN SEEBECK COEFFICIENTS

Based on the magnon bands [see Appendix B 2 (C 2) for the monolayer (bilayer)] and SAMs in Figs. 1, 2(a), and 2(b), we compute here the SSC by the linearized Boltzmann equation in the constant relaxation time approximation.

In the semiclassical picture, each magnon carries SAM ΔS_k and propagates with group velocity $\vec{v}_k$. A deviation of its occupation from thermal equilibrium therefore generates a net spin current. Let $n_k = n(\vec{r}, \vec{k}, t)$ be the nonequilibrium

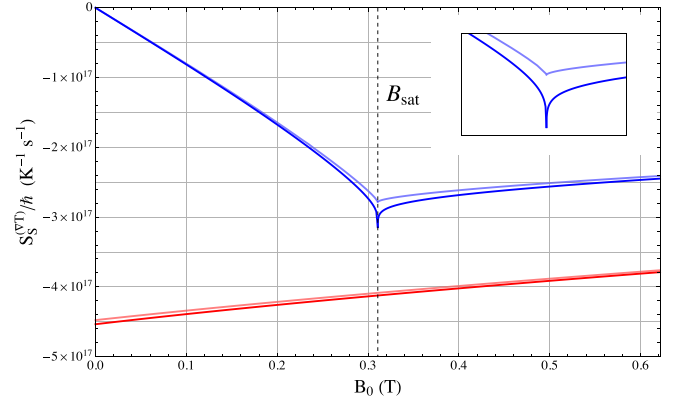


FIG. 3. Thermal SSC in a monolayer as a function of the external magnetic field B_0 . The transparent curves correspond to $S_S^{(\nabla T)}$ for all SAM equal $\hbar$. The red curves show $S_S^{(\nabla T)}$ along the easy axis for $\vec{B}_0 \parallel \hat{b}$, the blue curves along the intermediate axis in the canted ($B_0 < B_{\text{sat}}$) and saturated ($B_0 \geq B_{\text{sat}}$) phases for $\vec{B}_0 \parallel \hat{a}$. The inset highlights a zoomed-in region around the peak of $S_S^{(\nabla T)}$.

distribution function of the magnon occupation numbers. The magnon spin current [23] in 2D reads

$$\vec{J}_S = \frac{1}{(2\pi)^2} \int \Delta S_k \vec{v}_k (n_k - n_k^{(0)}) d^2k, \quad (18)$$

where ΔS_k the SAM from Sec. III, $\vec{v}_k = d\omega_{\vec{k}}/d\vec{k}$ the magnon group velocity and $n^{(0)} = (e^\sigma - 1)^{-1}$ the occupation number at thermal equilibrium from the Bose-Einstein distribution with $\sigma = \hbar\omega_{\vec{k}}/(k_B T)$. Substituting the magnon distribution function n_k obtained from the linearized Boltzmann equation ($n_k = n^{(0)} - \tau_k \vec{v}_k \cdot \nabla_r n_k$) for a constant relaxation time τ [23], the spin transport along η ($= x, z$) excited by a temperature gradient $\partial T/\partial \eta$ reads

$$\begin{aligned} J_S^{(\eta, \nabla T)} &= -\frac{\tau}{(2\pi)^2} \int \Delta S_k v_{k,\eta}^2 \frac{\partial n^{(0)}}{\partial T} d^2k \frac{\partial T}{\partial \eta} \\ &\equiv -S_S^{(\nabla T)}(B_0) \frac{\partial T}{\partial \eta}, \end{aligned} \quad (19)$$

where $S_S^{(\nabla T)}(B_0)$ is the thermal SSC due to thermal gradients. Likewise, Taylor expanding around $\varepsilon_{\vec{k}} (= \hbar\omega_{\vec{k}})$ for a small chemical potential μ in $n_\mu^{(0)} = (e^{\sigma_\mu} - 1)^{-1}$ with $\sigma_\mu = (\varepsilon_{\vec{k}} - \mu)/(k_B T)$, the spin transport excited by a chemical potential gradient $\partial \mu/\partial \eta$ reads

$$\begin{aligned} J_S^{(\eta, \nabla \mu)} &= -\frac{\tau}{(2\pi)^2} \int \Delta S_k v_{k,\eta}^2 \frac{\partial n_\mu^{(0)}}{\partial \mu} \bigg|_{\mu=0} d^2k \frac{\partial \mu}{\partial \eta} \\ &\equiv -S_S^{(\nabla \mu)}(B_0) \frac{\partial \mu}{\partial \eta}, \end{aligned} \quad (20)$$

where $S_S^{(\nabla \mu)}(B_0)$ is the diffusive SSC due to magnon accumulation. Since the magnon relaxation time in CrSBr is not known, we adopt a value $\tau \approx 10 \mu\text{s}$, which is typical for yttrium iron garnet films [35,36].

A. Monolayer CrSBr

Figure 3 shows the thermal SSC in the monolayer at $T = 5$ K. The red curve corresponds to magnetization and field

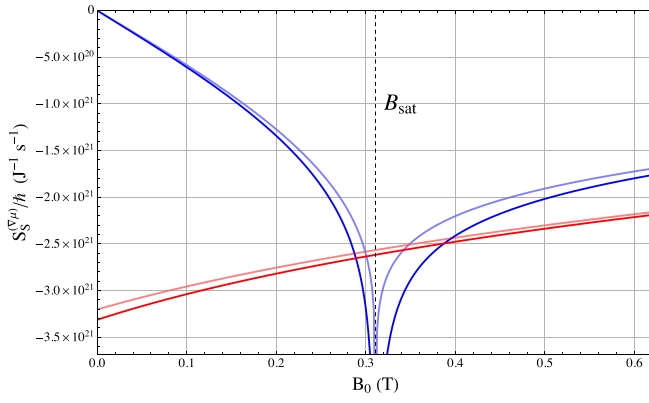


FIG. 4. Diffusive SSC in a monolayer as a function of the external magnetic field B_0 . The transparent curves correspond to $S_S^{(V\mu)}$ for all SAM equal $\hbar$. The red curves show $S_S^{(V\mu)}$ along the easy axis for $\vec{B}_0 \parallel \hat{b}$, the blue curves along the intermediate axis in the canted ($B_0 < B_{\text{sat}}$) and saturated ($B_0 \geq B_{\text{sat}}$) phases for $\vec{B}_0 \parallel \hat{a}$.

parallel with the easy axis (b). The dependence on magnetic field is weak because the magnon SAM is nearly constant and close to $\hbar$.

The blue curve corresponds to the external field along the intermediate axis (a), which causes a logarithmic divergence of the thermal SSC around the saturation field B_{sat} [21]. Imposing a constant $\Delta S = -\hbar$ (transparent curves) suppresses the enhancement at B_{sat} , where the curves meet linearly at a minimum and do not exhibit a peak. The predicted peak is, therefore, a spin-caloritronic signature of field-induced soft magnons. Figure 12 in Appendix D 1 illustrates the temperature dependence of the SSC at low temperatures, disregarding the temperature dependence of the magnetization.

Figure 4 shows the diffusive SSC in the monolayer at $T = 5$ K. The red curves correspond to magnetization and field parallel with the easy axis (b). The dependence on the external field is weak for the same reason as in the thermal SSC.

The blue curves correspond to the external field along the intermediate axis (a), which now causes an enhancement of an already present peak of the diffusive SSC. Even for fixed magnon spin $\Delta S = \hbar$, the diffusive SSC peaks near the softening field because the low-energy magnon occupation becomes highly sensitive to the chemical potential as the magnon gap closes. As $\omega \rightarrow 0$, the low-energy magnon occupation grows large compared to S . Consequently, higher-order magnon interactions neglected in linear spin wave theory become important and are expected to regularize the divergence.

Combining the nonuniversal SAM [Eq. (16)] with the transport equation (18) in the monolayer shows that the response to an easy-axis field is weak, whereas an intermediate-axis field produces a clear spin-caloritronic signature of magnon softening in the form of a measurable peak in the thermal SSC. This peak originates from the enhanced nonuniversal SAM and disappears when the spin is artificially fixed to $\hbar$. By contrast, the diffusive SSC already peaks because the low-energy occupation is highly sensitive to the chemical potential, with the nonuniversal SAM providing an additional enhancement.

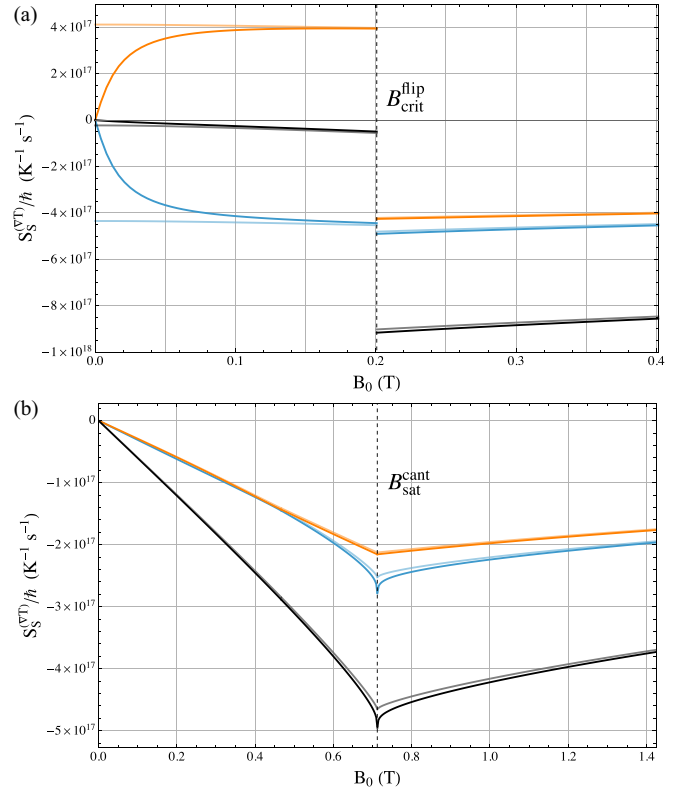


FIG. 5. Thermal SSC of the two magnon modes and their sum (black curves) in the bilayer as a function of the external magnetic field B_0 . The semi-transparent curves correspond to $S_S^{(VT)}$ with the SAM set to $\pm \hbar$ ($\hbar$) in the AFM (FM) phase. (a) $S_S^{(VT)}$ along the easy axis for $\vec{B}_0 \parallel \hat{b}$ in the AFM ($B_0 < B_{\text{crit}}^{\text{flip}}$) and FM ($B_0 \geq B_{\text{crit}}^{\text{flip}}$) phases. (b) $S_S^{(VT)}$ along the intermediate axis for $\vec{B}_0 \parallel \hat{a}$ in the canted ($B_0 < B_{\text{sat}}^{\text{cant}}$) and saturated ($B_0 \geq B_{\text{sat}}^{\text{cant}}$) phases.

Figure 13 in Appendix E 1 illustrates the temperature dependence at low temperatures. For completeness we show the magnon thermal conductivity for all phases in the monolayer in Fig. 16 in Appendix E 1.

B. Bilayer CrSBr

In this section, we discuss the SSC in the bilayer. Figure 5(a) shows the thermal SSC of the bilayer in the AFM and FM phases for thermal gradient and external magnetic field applied along the easy axis ($\vec{B}_0 \parallel \hat{b}$) at $T = 5$ K. For fields below, the spin-flip critical field $B_{\text{crit}}^{\text{flip}}$, the system is in the AFM phase. Both magnon branches contribute to the total thermal SSC proportional to their thermal occupation and partially cancel since they carry opposite SAM. Setting the SAM to $\pm \hbar$ for the two modes appears to be a bad approximation at weak magnetic fields, but the error largely cancels in the total thermal SSC (black curves) apart from giving a small nonzero value. Above $B_0 = B_{\text{crit}}^{\text{flip}}$ the sign of the SAM of both magnon branches is the same and close to $\hbar$ in both branches. The field dependence of the thermal SSC remains relatively weak. The thermal SSC of the bilayer in the AFM and FM phases shows a discontinuity at the spin-flip field, but no divergence.

Figure 5(b) shows the thermal SSC of the bilayer in the canted and saturated phases for thermal gradient and external

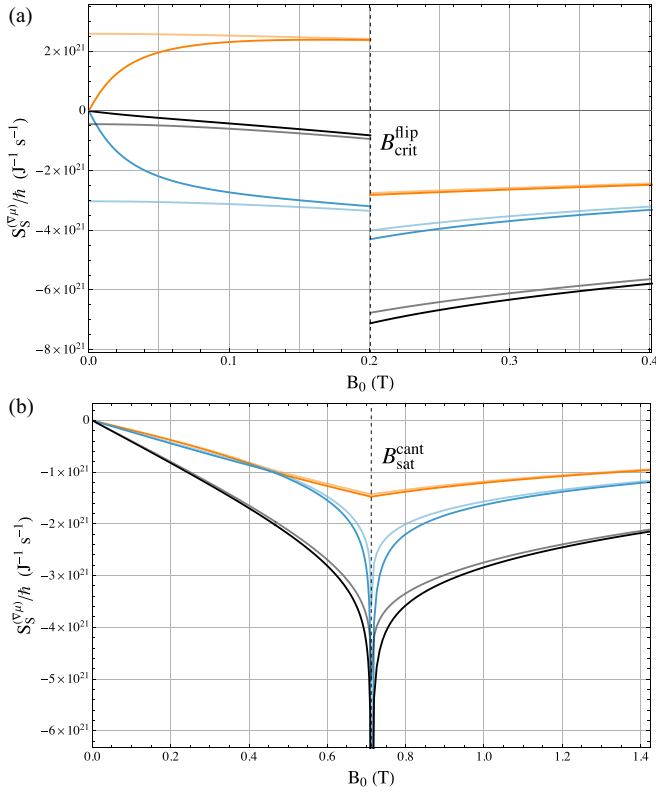


FIG. 6. Diffusive SSC of the two magnon modes and their sum (black curves) in the bilayer as a function of the external magnetic field B_0 . The semi-transparent curves correspond to $S_S^{(\nu\mu)}$ with the SAM set to $\pm\hbar$ ($\hbar$) in the AFM (FM) phase. (a) $S_S^{(\nu\mu)}$ along the easy axis for $\vec{B}_0 \parallel \hat{b}$ in the AFM ($B_0 < B_{crit}^{flip}$) and FM ($B_0 \geq B_{crit}^{flip}$) phases. (b) $S_S^{(\nu\mu)}$ along the intermediate axis for $\vec{B}_0 \parallel \hat{a}$ in the canted ($B_0 < B_{sat}^{cant}$) and saturated ($B_0 \geq B_{sat}^{cant}$) phases.

magnetic field applied along the intermediate axis ($\vec{B}_0 \parallel \hat{a}$) at $T = 5$ K. When the field approaches B_{sat}^{cant} , the frequency of lower mode approaches zero, leading to a divergence in the SAM. As in the monolayer, this strongly affects the thermal SSC since small- k magnons dominate spin transport. The thermal SSC of the high-frequency mode displays a pronounced kink at the phase transition. In the saturated phase ($B_0 \geq B_{sat}^{cant}$), the spins are forced to become parallel and the soft magnon mode hardens again. At high fields, the SAM of both modes approach $\hbar$. As with the canted phase in the monolayer, setting the SAM to $\hbar$ (semi-transparent curves) underestimates the thermal SSC and does not produce the peak at B_{sat}^{cant} . Figure 14 shows the total thermal SSC for various temperatures in all phases in the bilayer.

Figure 6(a) shows the diffusive SSC of the bilayer in the AFM and FM phases at $T = 5$ K. In both phases the diffusive SSC behaves much like the thermal one. For $B_0 < B_{crit}^{flip}$ the two magnon branches carry opposite SAM. Naively setting the SAM to $\pm\hbar$ overestimates the diffusive SSC which does not accurately go to zero for external fields near zero. Above the spin-flip transition both branches carry spin with the same sign and show a weak field dependence.

Figure 6(b) shows the diffusive SSC of the bilayer in the canted and saturated phases at $T = 5$ K. For $\vec{B}_0 \parallel \hat{a}$, the lower magnon mode softens as the external field approaches

B_{sat}^{cant} , producing a pronounced enhancement of the diffusive SSC peak. As in the monolayer, in contrast to the thermal SSC, the diffusive SSC exhibits a peak for the soft mode even when the SAM is fixed to $\Delta S = \hbar$ (semi-transparent curves). The nonuniversal SAM further amplifies this low-energy contribution. The relatively hard mode remains weakly field-dependent.

The bilayer SSC reflects the relative orientation of the two magnon branches and the resulting nonuniversal SAM. Along the easy axis, the opposite SAM of the AFM modes suppresses the total response at weak fields, while the spin-flip transition produces a discontinuity without a divergence. Along the intermediate axis, the soft lower mode generates pronounced thermal and diffusive SSC peaks near B_{sat}^{cant} ; the thermal peak is solely due to the nonuniversal SAM, whereas the diffusive peak is further amplified by it. These features provide experimentally accessible signatures of both the magnetic phase transitions and the renormalized SAM.

Figures 15 and 17 in Appendixes D 2 and E 2 show the total diffusive SSC and magnon thermal conductivity, respectively, for various temperatures in all phases in the bilayer.

V. CONCLUSION

We calculated the nonuniversal magnon SAM and the associated thermal and diffusive SSC for mono- and bilayer CrSBr, a particularly suitable platform with magnon modes in the GHz regime. The frequency of one of the two magnon modes in the antiferromagnet softens to zero under moderate in-plane magnetic fields perpendicular to the zero-field Neel vector, which leads to an algebraic divergence of the SAM and a logarithmic one in the thermal SSC. The latter allows for an experimentally accessible spin-caloritronic signature of soft magnons. In the diffusive SSC, the peak arising from the strong sensitivity of the response to the chemical potential at low frequencies is further enhanced by the nonuniversal SAM. In CrSBr, both triaxial anisotropy and intralayer dipole-dipole interactions produce the divergent SAM. More generally, our calculations indicate that either interaction is sufficient, so that the effects described here should occur in magnets, where one of them coexists with a soft magnon mode.

While our analysis focuses on bulk magnon transport, experiments are usually carried out electrically with heavy metal contacts to the vdW magnet. In contrast to Pt contacts on yttrium iron garnet films, the interface conductance that depends strongly on the degree of compensation [37] may play an important role.

Our results highlight the effect of nonuniversal magnon SAM in spin caloritronics. Future work should address how these bulk spin-caloritronic signatures are modified by interfacial effects, such as spin-mixing conductance [37,38], magnon-electron conversion efficiency [39], and spin backflow at metal-magnetic insulator interfaces [40]. Experiments on CrSBr and other vdW magnets are necessary to assess the relative importance of these contributions.

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DATA AVAILABILITY

The data supporting the findings of this article are openly available [41].

APPENDIX A: CRSBR MODEL AND MATERIAL VALUES

Figure 7 shows the CrSBr crystal structure and Table III lists the parameters used here.

APPENDIX B: MONOLAYER

1. Hamiltonian

The Hamiltonian for a monolayer reads [19]

$$H_A = - \underbrace{\sum_j \gamma \hbar \vec{B}_0 \cdot \vec{S}_{j,A}}_{H_{Ext}} - \underbrace{\sum_{j,\sigma} \mathcal{J}_\sigma \vec{S}_{j,A} \cdot \vec{S}_{j+\sigma,A}}_{H_{Ex}} - \underbrace{\sum_j [\mathcal{D}_x S_{x,j,A}^2 + \mathcal{D}_y S_{y,j,A}^2 + \mathcal{D}_z S_{z,j,A}^2]}_{H_{An}}, \quad (B1)$$

to which we add $H_{dip,A}$ from Eq. (2).

2. Resonance frequencies

The resonance frequencies for all phases in the monolayer have been calculated in Ref. [19] and are shown in Fig. 8.

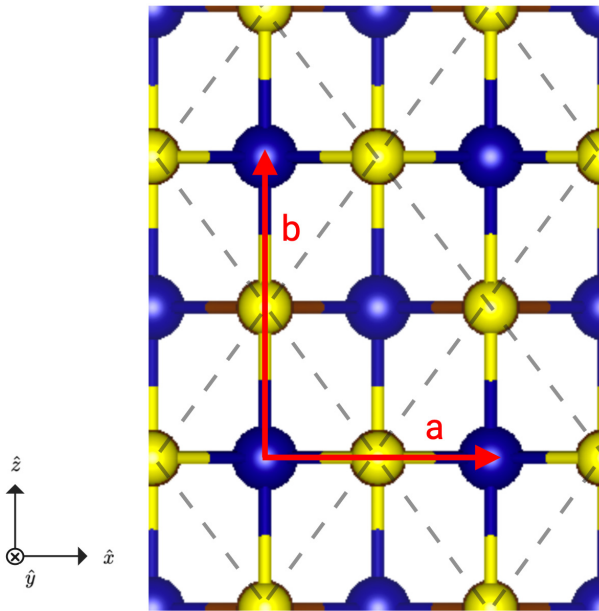


FIG. 7. Top view of monolayer CrSBr, illustrating the crystallographic axes and lattice vectors. Primitive unit cell in dashed lines. The magnetic chromium atoms are colored blue.

TABLE III. In-plane lattice parameters a and b , exchange interactions $\mathcal{J}_i$ [42], and anisotropy coefficients $\mathcal{D}_i$ [16].

a	3.54	10^{-10}m
b	4.73	10^{-10}m
$\mathcal{J}_1$	3.54	meV
$\mathcal{J}_2$	3.08	meV
$\mathcal{J}_3$	4.15	meV
$\mathcal{J}_\perp$	-15.5	μeV
$\mathcal{D}_x$	-12	μeV
$\mathcal{D}_y$	-78	μeV
$\mathcal{D}_z$		μeV

3. Canted configuration

Here, we describe the coordinate transformations when the spin texture is noncollinear to an anisotropy axis. The canted phase in the monolayer is described in the basis of $[\hat{e}_\alpha^A, \hat{e}_\beta^A, \hat{e}_\gamma^A]$ with transformations

$$\begin{aligned} \hat{x} &= \cos(\theta) \hat{e}_\alpha^A + \sin(\theta) \hat{e}_\gamma^A, \\ \hat{y} &= \hat{e}_\beta^A, \\ \hat{z} &= -\sin(\theta) \hat{e}_\alpha^A + \cos(\theta) \hat{e}_\gamma^A, \end{aligned} \quad (B2)$$

as illustrated in Fig. 9.

4. Eigenvalue matrices

Here, we show the matrix elements of $\mathcal{H}^{(l)}$ from Eq. (4) for all phases in the monolayer. The elements for the external field along the easy axis (b) in the monolayer read

$$\begin{aligned} A_k^{(m_{\text{FM}})} &= 2S\gamma(k) + S(\mathcal{D}_x + \mathcal{D}_y) - \hbar\gamma B_0 - 2S(\mathcal{J} + \mathcal{D}_z) \\ &\quad - \frac{1}{2}\mu_0\gamma\hbar M_s f(k) - \frac{1}{2}\mu_0\gamma\hbar M_s \frac{k_x^2}{k^2} (1 - f(k)), \\ B_k^{(m_{\text{FM}})} &= S(\mathcal{D}_x - \mathcal{D}_y) + \frac{1}{2}\mu_0\gamma\hbar M_s f(k) \\ &\quad - \frac{1}{2}\mu_0\gamma\hbar M_s \frac{k_x^2}{k^2} (1 - f(k)). \end{aligned} \quad (B3)$$

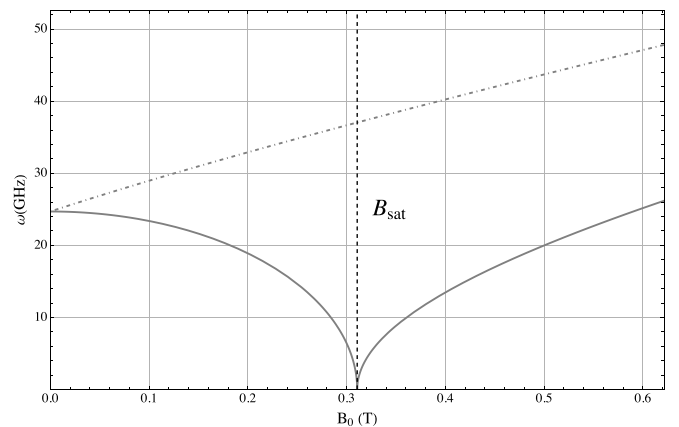


FIG. 8. Resonance frequencies ($k = 0$) in the monolayer. The dash-dotted curve corresponds to $\vec{B}_0 \parallel \hat{z}$ ($\hat{b}$), the solid curve corresponds to $\vec{B}_0 \parallel \hat{x}$ ($\hat{a}$).

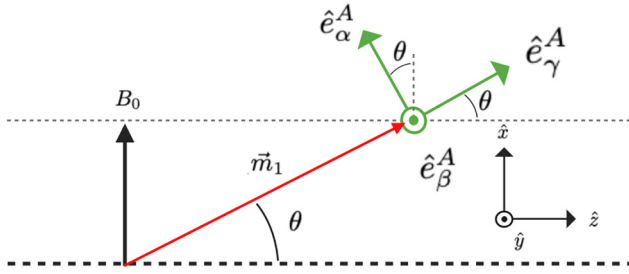


FIG. 9. Canted configuration of magnetic moments in the monolayer for $\vec{B}_0 \parallel \hat{x}$ ($\hat{a}$), showing the local rotated reference frame and the corresponding unit vectors.

The elements for the canted phase in the monolayer read

$$\begin{aligned}
 A_k^{(m_{\text{cant}})} &= 2S\gamma(k) + S(\mathcal{D}_x \cos^2(\theta) + \mathcal{D}_z \sin^2(\theta) + \mathcal{D}_y) \\
 &\quad - \hbar\gamma B_0 \sin(\theta) - 2S(\mathcal{J} + \mathcal{D}_x \sin^2(\theta) + \mathcal{D}_z \cos^2(\theta)) \\
 &\quad - \frac{1}{2}\mu_0\gamma\hbar M_s f(k) - \frac{1}{2}\mu_0\gamma\hbar M_s \frac{k_\alpha^2}{k^2}(1-f(k)), \\
 B_k^{(m_{\text{cant}})} &= S(\mathcal{D}_x \cos^2(\theta) + \mathcal{D}_z \sin^2(\theta) - \mathcal{D}_y) \\
 &\quad + \frac{1}{2}\mu_0\gamma\hbar M_s f(k) - \frac{1}{2}\mu_0\gamma\hbar M_s \frac{k_\alpha^2}{k^2}(1-f(k)).
 \end{aligned} \tag{B4}$$

The elements for the saturated phase in the monolayer read

$$\begin{aligned}
 A_k^{(m_{\text{sat}})} &= 2S\gamma(k) + S(\mathcal{D}_z + \mathcal{D}_y) - \hbar\gamma B_0 - 2S(\mathcal{J} + \mathcal{D}_x) \\
 &\quad - \frac{1}{2}\mu_0\gamma\hbar M_s f(k) - \frac{1}{2}\mu_0\gamma\hbar M_s \frac{k_z^2}{k^2}(1-f(k)), \\
 B_k^{(m_{\text{sat}})} &= S(\mathcal{D}_z - \mathcal{D}_y) + \frac{1}{2}\mu_0\gamma\hbar M_s f(k) \\
 &\quad - \frac{1}{2}\mu_0\gamma\hbar M_s \frac{k_z^2}{k^2}(1-f(k)).
 \end{aligned} \tag{B5}$$

$k_{\alpha,i}$ ($i = A, B$) is related to k_x and k_z through Eq. (B2). In all phases,

$$\begin{aligned}
 \gamma(k) &= 2\mathcal{J}_1 \cos(k_x a) + 2\mathcal{J}_3 \cos(k_z b) \\
 &\quad + 2\mathcal{J}_2 \left(\cos\left(\frac{1}{2}k_x a + \frac{1}{2}k_z b\right) + \cos\left(\frac{1}{2}k_x a - \frac{1}{2}k_z b\right) \right),
 \end{aligned} \tag{B6}$$

and

$$\mathcal{J} = 2\mathcal{J}_1 + 4\mathcal{J}_2 + 2\mathcal{J}_3. \tag{B7}$$

APPENDIX C: BILAYER

1. Hamiltonian

The Hamiltonian for a bilayer reads [19]

$$H_{Bi} = H_A + H_B + H_{\text{int}}, \tag{C1}$$

where

$$H_{\text{int}} = - \sum_j \mathcal{J}_\perp \vec{S}_{j,A} \cdot \vec{S}_{j,B}, \tag{C2}$$

to which we add $H_{\text{dip},A}$ and $H_{\text{dip},B}$ from Eq. (2).

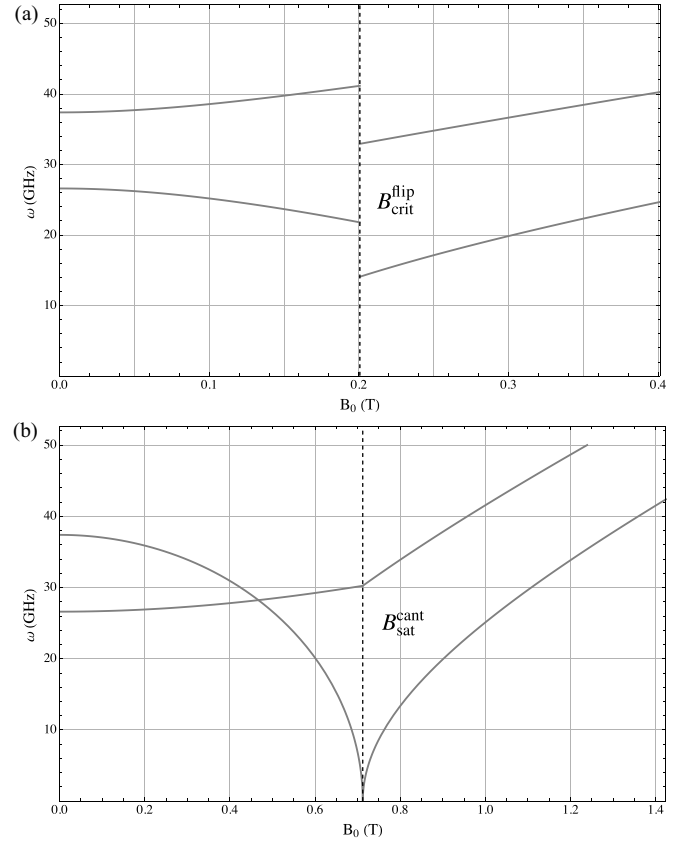


FIG. 10. Resonance frequencies ($k = 0$) in the bilayer. (a) AFM ($\vec{B}_0 < B_{\text{crit}}^{\text{flip}}$) and FM ($\vec{B}_0 \geq B_{\text{crit}}^{\text{flip}}$) phases with $\vec{B}_0 \parallel \hat{z}$ ($\hat{b}$). (b) Canted ($\vec{B}_0 < B_{\text{sat}}^{\text{cant}}$) and saturated ($\vec{B}_0 \geq B_{\text{sat}}^{\text{cant}}$) phases with $\vec{B}_0 \parallel \hat{x}$ ($\hat{a}$).

2. (A)FMR

The resonance frequencies for all phases in the bilayer have been calculated in Ref. [19] and are shown in Fig. 10.

3. Canted configuration

Here, we describe the coordinate transformations when the spin texture is noncollinear to an anisotropy axis. The canted phase in the bilayer is described in the basis of $[\hat{e}_\alpha^A, \hat{e}_\beta^A, \hat{e}_\gamma^A]$ and $[\hat{e}_\alpha^B, \hat{e}_\beta^B, \hat{e}_\gamma^B]$ with transformations

$$\begin{aligned}
 \hat{x} &= \cos(\theta)\hat{e}_\alpha^A + \sin(\theta)\hat{e}_\gamma^A, \\
 \hat{y} &= \hat{e}_\beta^A, \\
 \hat{z} &= -\sin(\theta)\hat{e}_\alpha^A + \cos(\theta)\hat{e}_\gamma^A,
 \end{aligned} \tag{C3}$$

and

$$\begin{aligned}
 \hat{x} &= -\cos(\theta)\hat{e}_\alpha^B + \sin(\theta)\hat{e}_\gamma^B, \\
 \hat{y} &= \hat{e}_\beta^B, \\
 \hat{z} &= -\sin(\theta)\hat{e}_\alpha^B - \cos(\theta)\hat{e}_\gamma^B,
 \end{aligned} \tag{C4}$$

as illustrated in Fig. 11.

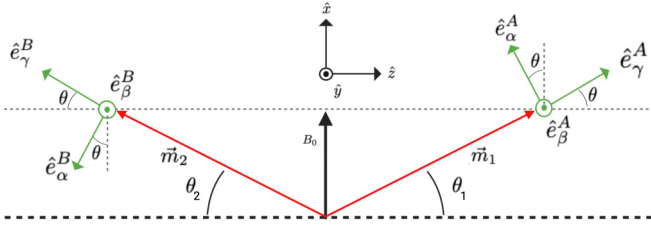


FIG. 11. Canted configuration of magnetic moments in the bilayer for $\vec{B}_0 \parallel \hat{x}$ ($\hat{a}$), showing the local rotated reference frames and the corresponding unit vectors.

4. Eigenvalue matrix

Here, we show the matrix $\mathcal{H}^{(l)}$ for all phases in the bilayer. The matrix for the AFM phase in the bilayer reads

$$\mathcal{H}^{(b_{AFM})} = \begin{bmatrix} -A_k^{A,\parallel z} & 0 & -B_k^{A,\parallel z} & \mathcal{J}_\perp S \\ 0 & -A_k^{B,\parallel -z} & \mathcal{J}_\perp S & -B_k^{A,\parallel z} \\ -B_k^{A,\parallel z} & \mathcal{J}_\perp S & -A_k^{A,\parallel z} & 0 \\ \mathcal{J}_\perp S & -B_k^{A,\parallel z} & 0 & -A_k^{B,\parallel -z} \end{bmatrix}, \quad (C5)$$

where

$$\begin{aligned} A_k^{A,\parallel z} &= 2S\gamma(k) + S(\mathcal{D}_x + \mathcal{D}_y) - \hbar\gamma B_0 - 2S(\mathcal{J} + \mathcal{D}_z) \\ &\quad + S\mathcal{J}_\perp - \frac{1}{2}\mu_0\gamma\hbar M_s f(k) - \frac{1}{2}\mu_0\gamma\hbar M_s \frac{k_x^2}{k^2}(1-f(k)), \\ A_k^{B,\parallel -z} &= 2S\gamma(k) + S(\mathcal{D}_x + \mathcal{D}_y) + \hbar\gamma B_0 - 2S(\mathcal{J} + \mathcal{D}_z) \\ &\quad + S\mathcal{J}_\perp - \frac{1}{2}\mu_0\gamma\hbar M_s f(k) - \frac{1}{2}\mu_0\gamma\hbar M_s \frac{k_x^2}{k^2}(1-f(k)) \\ &\quad + \frac{1}{2}\mu_0\gamma\hbar M_s f(k) - \frac{1}{2}\mu_0\gamma\hbar M_s \frac{k_x^2}{k^2}(1-f(k)), \\ B_k^{A,\parallel z} &= S(\mathcal{D}_x - \mathcal{D}_y) + \frac{1}{2}\mu_0\gamma\hbar M_s f(k) \\ &\quad - \frac{1}{2}\mu_0\gamma\hbar M_s \frac{k_x^2}{k^2}(1-f(k)). \end{aligned} \quad (C6)$$

The superscripts indicate the magnetization of sublattice A (B) is along z ($-z$).

The matrix for the FM phase in the bilayer reads

$$\mathcal{H}^{(b_{FM})} = \begin{bmatrix} -A_k^{FM} & -\mathcal{J}_\perp S & -B_k^{FM} & 0 \\ -\mathcal{J}_\perp S & -A_k^{FM} & 0 & -B_k^{FM} \\ -B_k^{FM} & 0 & -A_k^{FM} & -\mathcal{J}_\perp S \\ 0 & -B_k^{FM} & -\mathcal{J}_\perp S & -A_k^{FM} \end{bmatrix}, \quad (C7)$$

where

$$\begin{aligned} A_k^{FM} &= 2S\gamma(k) + S(\mathcal{D}_x + \mathcal{D}_y) - \hbar\gamma B_0 \\ &\quad - 2S(\mathcal{J} + \mathcal{D}_z) - S\mathcal{J}_\perp \\ &\quad - \frac{1}{2}\mu_0\gamma\hbar M_s f(k) - \frac{1}{2}\mu_0\gamma\hbar M_s \frac{k_x^2}{k^2}(1-f(k)), \\ B_k^{FM} &= S(\mathcal{D}_x - \mathcal{D}_y) + \frac{1}{2}\mu_0\gamma\hbar M_s f(k) \\ &\quad - \frac{1}{2}\mu_0\gamma\hbar M_s \frac{k_x^2}{k^2}(1-f(k)). \end{aligned} \quad (C8)$$

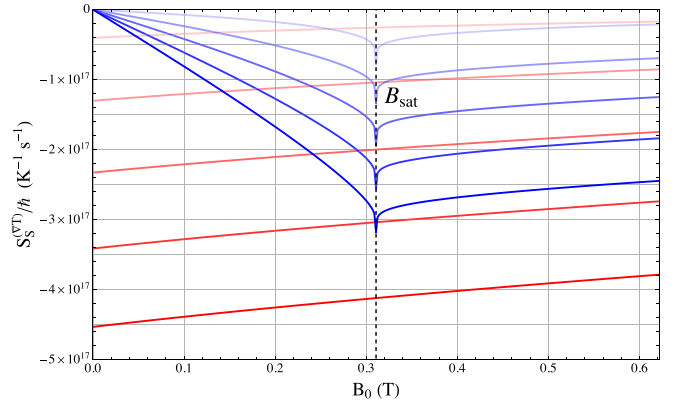


FIG. 12. Thermal SSC for various temperatures in a monolayer as a function of the external magnetic field B_0 . The red curves show $S_S^{(VT)}$ along the easy axis for $\vec{B}_0 \parallel \hat{b}$, the blue curves along the intermediate axis in the canted ($B_0 < B_{\text{sat}}$) and saturated ($B_0 \geq B_{\text{sat}}$) phases for $\vec{B}_0 \parallel \hat{a}$. The temperature ranges from $T = 1$ K (most transparent curve) to $T = 5$ K (opaque curve) in steps of 1 K.

The matrix for the canted phase in the bilayer reads

$$\mathcal{H}^{(b_{\text{cant}})} = \begin{bmatrix} -A_k^{A,\parallel \gamma^A} & -M^+ & -B_k^{A,\parallel \gamma^A} & -M^- \\ -M^+ & -A_k^{B,\parallel \gamma^B} & -M^- & -B_k^{B,\parallel \gamma^B} \\ -B_k^{A,\parallel \gamma^A} & -M^- & -A_k^{A,\parallel \gamma^A} & -M^+ \\ -M^- & -B_k^{B,\parallel \gamma^B} & -M^+ & -A_k^{B,\parallel \gamma^B} \end{bmatrix}, \quad (C9)$$

where

$$\begin{aligned} A_k^{A,\parallel \gamma^A} &= 2S\gamma(k) + S(\mathcal{D}_x \cos^2(\theta) + \mathcal{D}_z \sin^2(\theta) + \mathcal{D}_y) \\ &\quad - \hbar\gamma B_0 \sin(\theta) - 2S(\mathcal{J} + \mathcal{D}_x \sin^2(\theta) + \mathcal{D}_z \cos^2(\theta)) \\ &\quad + \cos(2\theta)S\mathcal{J}_\perp - \frac{1}{2}\mu_0\gamma\hbar M_s f(k) \\ &\quad - \frac{1}{2}\mu_0\gamma\hbar M_s \frac{k_{\alpha A}^2}{k^2}(1-f(k)), \end{aligned}$$

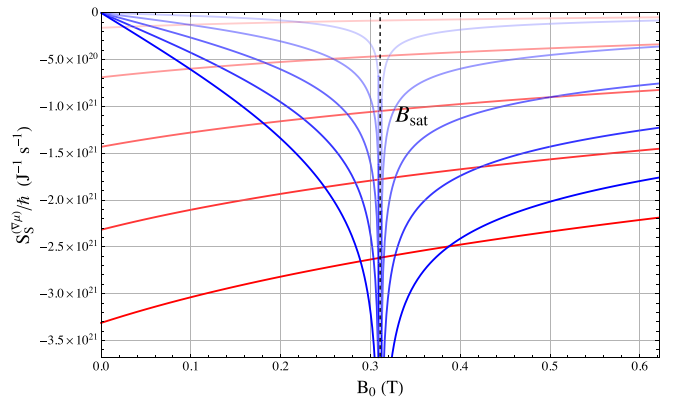


FIG. 13. Diffusive SSC for various temperatures in a monolayer as a function of the external magnetic field B_0 . The red curves show $S_S^{(V\mu)}$ along the easy axis for $\vec{B}_0 \parallel \hat{b}$, the blue curves along the intermediate axis in the canted ($B_0 < B_{\text{sat}}$) and saturated ($B_0 \geq B_{\text{sat}}$) phases for $\vec{B}_0 \parallel \hat{a}$. The temperature ranges from $T = 1$ K (most transparent curve) to $T = 5$ K (opaque curve) in steps of 1 K.

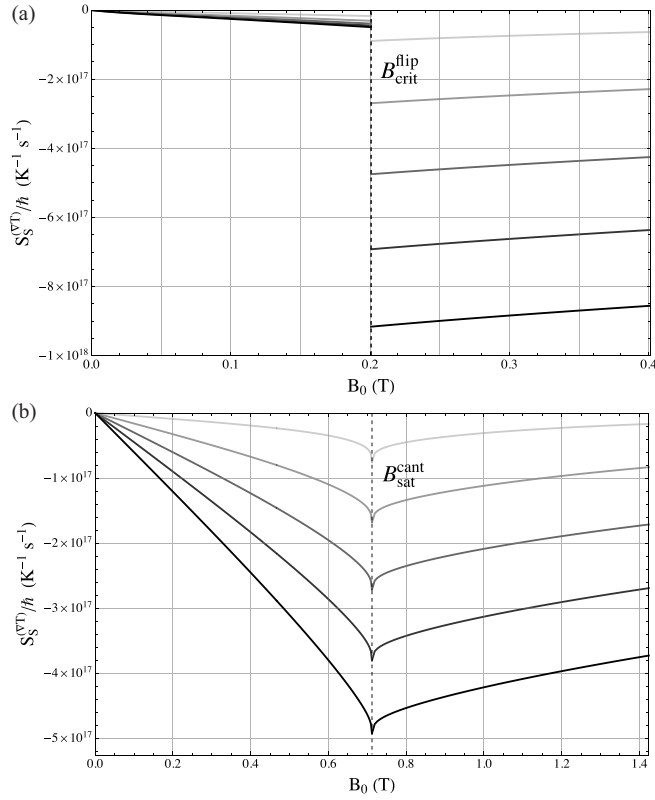


FIG. 14. Thermal SSC of the sum of the two magnon modes for various temperatures in the bilayer as a function of the external magnetic field B_0 . The temperature ranges from $T = 1$ K (most transparent curves) to $T = 5$ K (opaque curves) in steps of 1 K. (a) $S_S^{(VT)}$ along the easy axis for $\vec{B}_0 \parallel \hat{b}$ in the AFM ($B_0 < B_{crit}^{flip}$) and FM ($B_0 \geq B_{crit}^{flip}$) phases. (b) $S_S^{(VT)}$ along the intermediate axis for $\vec{B}_0 \parallel \hat{a}$ in the canted ($B_0 < B_{sat}^{cant}$) and saturated ($B_0 \geq B_{sat}^{cant}$) phases.

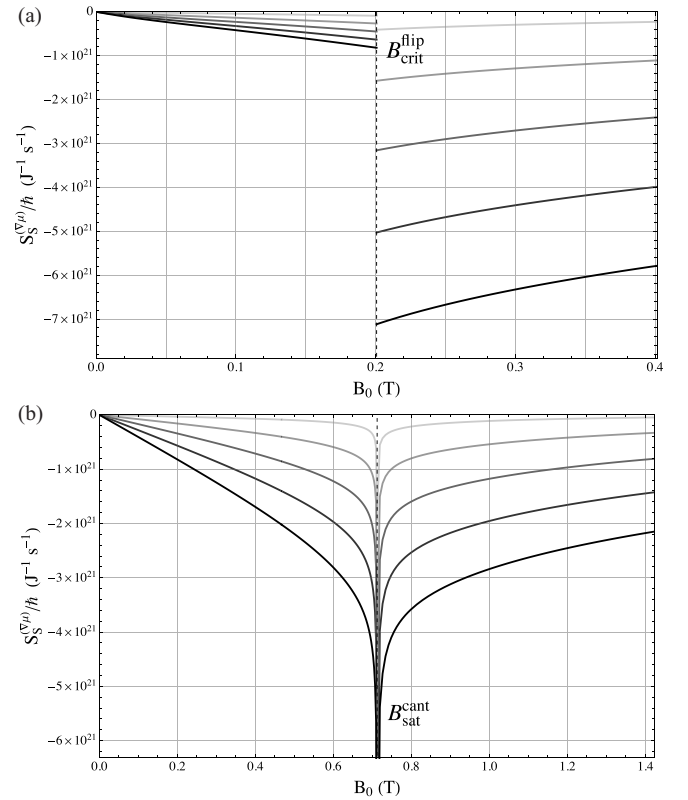


FIG. 15. Diffusive SSC of the sum of the two magnon modes for various temperatures in the bilayer as a function of the external magnetic field B_0 . The temperature ranges from $T = 1$ K (most transparent curves) to $T = 5$ K (opaque curves) in steps of 1 K. (a) $S_S^{(V\mu)}$ along the easy axis for $\vec{B}_0 \parallel \hat{b}$ in the AFM ($B_0 < B_{crit}^{flip}$) and FM ($B_0 \geq B_{crit}^{flip}$) phases. (b) $S_S^{(V\mu)}$ along the intermediate axis for $\vec{B}_0 \parallel \hat{a}$ in the canted ($B_0 < B_{sat}^{cant}$) and saturated ($B_0 \geq B_{sat}^{cant}$) phases.

$$\begin{aligned}
 A_k^{B, \parallel \gamma^B} &= 2S\gamma(k) + S(\mathcal{D}_x \cos^2(\theta) + \mathcal{D}_z \sin^2(\theta) + \mathcal{D}_y) \\
 &\quad - \hbar\gamma B_0 \sin(\theta) - 2S(\mathcal{J} + \mathcal{D}_x \sin^2(\theta) + \mathcal{D}_z \cos^2(\theta)) \\
 &\quad + \cos(2\theta)S\mathcal{J}_\perp \\
 &\quad - \frac{1}{2}\mu_0\gamma\hbar M_s f(k) - \frac{1}{2}\mu_0\gamma\hbar M_s \frac{k_{\alpha,B}^2}{k^2} (1 - f(k)), \\
 B_k^{A, \parallel \gamma^A} &= S(\mathcal{D}_x \cos^2(\theta) + \mathcal{D}_z \sin^2(\theta) - \mathcal{D}_y) \\
 &\quad + \frac{1}{2}\mu_0\gamma\hbar M_s f(k) - \frac{1}{2}\mu_0\gamma\hbar M_s \frac{k_{\alpha,A}^2}{k^2} (1 - f(k)), \\
 B_k^{B, \parallel \gamma^B} &= S(\mathcal{D}_x \cos^2(\theta) + \mathcal{D}_z \sin^2(\theta) - \mathcal{D}_y) \\
 &\quad + \frac{1}{2}\mu_0\gamma\hbar M_s f(k) - \frac{1}{2}\mu_0\gamma\hbar M_s \frac{k_{\alpha,B}^2}{k^2} (1 - f(k)), \\
 M^+ &= \frac{1}{2}\mathcal{J}_\perp S(1 - \cos(2\theta)), \\
 M^- &= -\frac{1}{2}\mathcal{J}_\perp S(1 + \cos(2\theta)). \tag{C10}
 \end{aligned}$$

The superscripts indicate the magnetization of sublattice A (B) is along $\hat{e}_\gamma^A$ ($\hat{e}_\gamma^B$), as shown in Fig. 11.

The matrix for the saturated phase in the bilayer reads

$$\mathcal{H}^{(b_{sat})} = \begin{bmatrix} -A_k^{sat} & -\mathcal{J}_\perp S & -B_k^{sat} & 0 \\ -\mathcal{J}_\perp S & -A_k^{sat} & 0 & -B_k^{sat} \\ -B_k^{sat} & 0 & -A_k^{sat} & -\mathcal{J}_\perp S \\ 0 & -B_k^{sat} & -\mathcal{J}_\perp S & -A_k^{sat} \end{bmatrix}, \tag{C11}$$

where

$$\begin{aligned}
 A_k^{sat} &= 2S\gamma(k) + S(\mathcal{D}_z + \mathcal{D}_y) - \hbar\gamma B_0 \\
 &\quad - 2S(\mathcal{J} + \mathcal{D}_x) - S\mathcal{J}_\perp \\
 &\quad - \frac{1}{2}\mu_0\gamma\hbar M_s f(k) - \frac{1}{2}\mu_0\gamma\hbar M_s \frac{k_z^2}{k^2} (1 - f(k)), \\
 B_k^{sat} &= S(\mathcal{D}_z - \mathcal{D}_y) + \frac{1}{2}\mu_0\gamma\hbar M_s f(k) \\
 &\quad - \frac{1}{2}\mu_0\gamma\hbar M_s \frac{k_z^2}{k^2} (1 - f(k)). \tag{C12}
 \end{aligned}$$

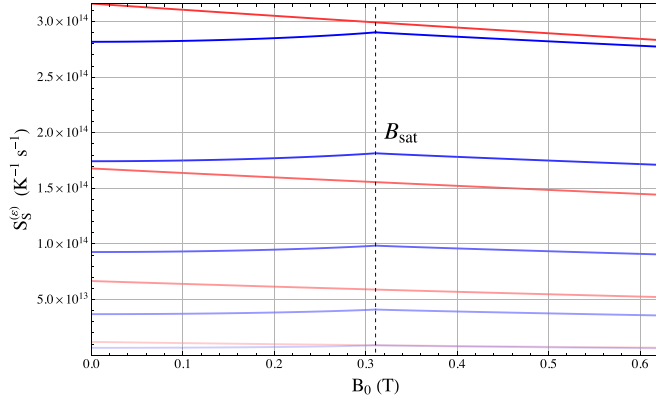


FIG. 16. Magnon thermal conductivity for various temperatures in a monolayer as a function of the external magnetic field B_0 . The temperature ranges from $T = 1$ K (most transparent curves) to $T = 5$ K (opaque curves) in steps of 1 K. The red curves show κ for $\vec{B}_0 \parallel \hat{b}$, the blue curves in the canted ($B_0 < B_{\text{sat}}$) and saturated ($B_0 \geq B_{\text{sat}}$) phases for $\vec{B}_0 \parallel \hat{a}$.

and $k_{\alpha,i}$ ($i = A, B$) is related to k_x and k_z through Eqs. (C3) and (C4). In all phases,

$$\begin{aligned} \gamma(k) = & 2\mathcal{J}_1 \cos(k_x a) + 2\mathcal{J}_3 \cos(k_z b) \\ & + 2\mathcal{J}_2 \left(\cos\left(\frac{1}{2}k_x a + \frac{1}{2}k_z b\right) \right. \\ & \left. + \cos\left(\frac{1}{2}k_x a - \frac{1}{2}k_z b\right) \right), \end{aligned} \quad (\text{C13})$$

and

$$\mathcal{J} = 2\mathcal{J}_1 + 4\mathcal{J}_2 + 2\mathcal{J}_3. \quad (\text{C14})$$

APPENDIX D: SSC AT VARIOUS TEMPERATURES

1. Monolayer

Figure 12 shows the thermal SSC in the monolayer with temperatures ranging from $T = 1$ K (most transparent curve) to $T = 5$ K (opaque curve) in steps of 1 K.

Figure 13 shows the diffusive SSC in the monolayer with temperatures ranging from $T = 1$ K (most transparent curve) to $T = 5$ K (opaque curve) in steps of 1 K.

2. Bilayer

Figures 14(a) (AFM-FM) and 14(b) (canted-saturated) show the thermal SSC in the bilayer with temperatures ranging from $T = 1$ K (most transparent curves) to $T = 5$ K (opaque curves) in steps of 1 K.

Figures 15(a) (AFM-FM) and 15(b) (canted-saturated) show the diffusive SSC in the bilayer with temperatures ranging from $T = 1$ K (most transparent curves) to $T = 5$ K (opaque curves) in steps of 1 K.

APPENDIX E: MAGNON THERMAL CONDUCTIVITY

Here, we show the magnon thermal conductivity in the mono- and bilayer. The heat flux flowing in the η direction

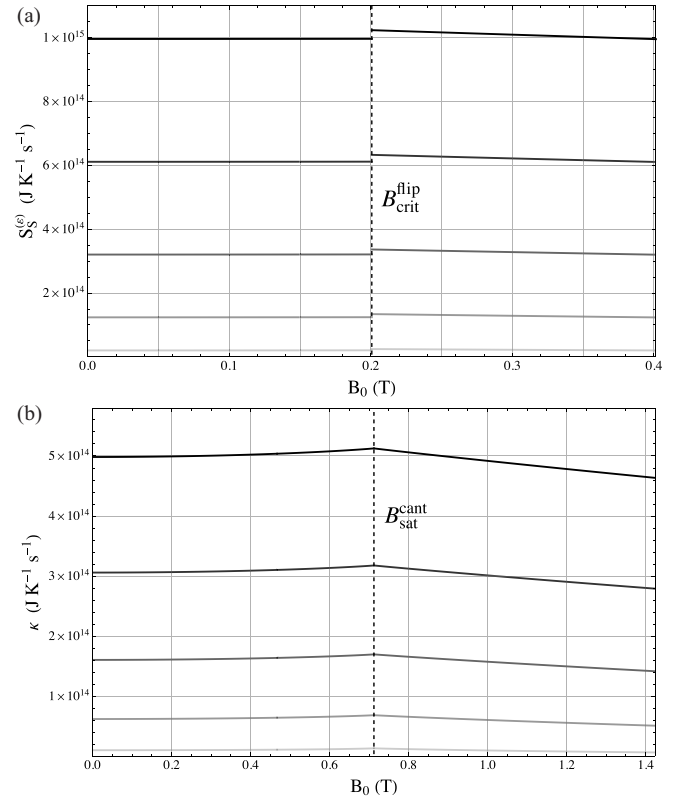


FIG. 17. Magnon thermal conductivity of the sum of the two magnon modes for various temperatures in the bilayer as a function of the external magnetic field B_0 . The temperature ranges from $T = 1$ K (most transparent curves) to $T = 5$ K (opaque curves) in steps of 1 K. (a) κ for $\vec{B}_0 \parallel \hat{b}$ in the AFM ($B_0 < B_{\text{crit}}^{\text{flip}}$) and FM ($B_0 \geq B_{\text{crit}}^{\text{flip}}$) phases. (b) κ for $\vec{B}_0 \parallel \hat{a}$ in the canted ($B_0 < B_{\text{sat}}^{\text{cant}}$) and saturated ($B_0 \geq B_{\text{sat}}^{\text{cant}}$) phases.

is

$$\begin{aligned} J_S^{(\eta, \varepsilon)} &= -\frac{\tau}{(2\pi)^2} \int \hbar \omega_{\vec{k}} v_{\vec{k}, \eta}^2 \frac{\partial n^0}{\partial T} d^2 k \frac{\partial T}{\partial \eta} \\ &\equiv -\kappa(B_0) \frac{\partial T}{\partial \eta}, \end{aligned} \quad (\text{E1})$$

where we introduced the magnon thermal conductivity $\kappa(B_0)$.

1. Monolayer

Figure 16 shows the magnon thermal conductivity in the monolayer with temperatures ranging from $T = 1$ K (most transparent curve) to $T = 5$ K (opaque curve) in steps of 1 K.

2. Bilayer

Figures 17(a) (AFM-FM) and 17(b) (canted-saturated) show the magnon thermal conductivity in the bilayer with temperatures ranging from $T = 1$ K (most transparent curves) to $T = 5$ K (opaque curves) in steps of 1 K.

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