Supplemental Material: All-Optical Generation of Antiferromagnetic Magnon Currents via the Spin Circular Photogalvanic Effect

SPIN HAMILTONIAN IN THE MAGNON BASIS

In this Section we provide details on the spin Hamiltonian in the magnon basis, the spin current operator and the interaction Hamiltonian for stimulated Raman scattering. We consider a general spin Hamiltonian

$$H = \sum_{\langle ij \rangle} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + J_z \sum_{\langle ij \rangle} S_i^z S_j^z - g\mu_B \mathbf{B}_0 \cdot \sum_i \mathbf{S}_i + \sum_{\langle ij \rangle} D\nu_{ij} \hat{\mathbf{z}} \cdot (\mathbf{S}_i \times \mathbf{S}_j), \quad (1)$$

where J_{ij} are bilinear exchange interactions, J_z is an easy-axis anisotropy, and $\mathbf{B}_0 = B_0\hat{\mathbf{z}}$ is an external magnetic field along the easy axis. Also, D is the next-nearest neighbor Dzyaloshinskii-Moriya interaction (DMI) strength (the nearest neighbor DMI vanishes by symmetry), and the function ν_{ij} encodes the phase accumulated by electrons hopping along isosceles triangles, which is $\nu_{ij} = \pm 1$ for hopping in a clockwise (anticlockwise) direction. The low-energy excitations of H are found to lowest order in $(1/S)$ by a Holstein-Primakoff transformation, and transforming to Fourier space the Hamiltonian is of the form

$$H = S \sum_{\mathbf{k}} \Psi_{\mathbf{k}}^\dagger H_{\mathbf{k}} \Psi_{\mathbf{k}}, \quad (2)$$

where $\Psi_{\mathbf{k}}^\dagger = (a_{\mathbf{k}}^\dagger, b_{-\mathbf{k}})$ is a Nambu spinor and $H_{\mathbf{k}} = h_0 \mathbf{1} + \mathbf{h} \cdot \boldsymbol{\tau}$, with $\boldsymbol{\tau}$ the vector of Pauli matrices. The components of the Hamiltonian are given by

$$\begin{aligned} h_0 &= J + 2J_2 \sum_i \cos(\mathbf{k} \cdot \boldsymbol{\delta}_i^{(2)}) \quad (3) \\ h_x &= \sum_i [J_1 \cos(\mathbf{k} \cdot \boldsymbol{\delta}_i^{(1)}) + J_3 \cos(\mathbf{k} \cdot \boldsymbol{\delta}_i^{(3)})] \\ h_y &= \sum_i [J_1 \sin(\mathbf{k} \cdot \boldsymbol{\delta}_i^{(1)}) + J_3 \sin(\mathbf{k} \cdot \boldsymbol{\delta}_i^{(3)})] \\ h_z &= \frac{B}{S} + 2D \sum_i \sin(\mathbf{k} \cdot \boldsymbol{\delta}_i^{(2)}), \end{aligned}$$

where $J = 3J_1 - 6J_2 + 3J_3 + 3J_z$, $B = g\mu_B B_0$, and $\boldsymbol{\delta}_i^{(n)}$ are the nth nearest neighbor vectors of the lattice. The Hamiltonian is diagonalized to $H = \sum_{\mathbf{k}} \epsilon_{\alpha\mathbf{k}} \alpha_{\mathbf{k}}^\dagger \alpha_{\mathbf{k}} + \epsilon_{\beta\mathbf{k}} \beta_{-\mathbf{k}}^\dagger \beta_{-\mathbf{k}}$ via a paraunitary matrix $U_{\mathbf{k}}$, given by

$$U_{\mathbf{k}} = \begin{pmatrix} \cosh(\theta_{\mathbf{k}}/2) & e^{i\phi_{\mathbf{k}}} \sinh(\theta_{\mathbf{k}}/2) \\ e^{-i\phi_{\mathbf{k}}} \sinh(\theta_{\mathbf{k}}/2) & \cosh(\theta_{\mathbf{k}}/2) \end{pmatrix}. \quad (4)$$

Here the Bogoliubov angles are given by $\phi_{\mathbf{k}} = -\arctan(h_y/h_x)$ and $\theta_{\mathbf{k}} = -\arctan(\sqrt{h_x^2 + h_y^2}/h_0)$ [1].

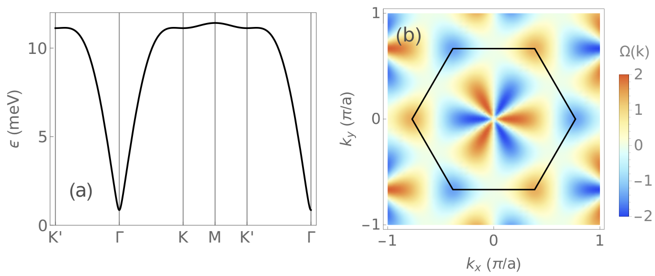

FIG. 1. **Bandstructure and Berry curvature.** (a) Magnon band structure of a collinear honeycomb antiferromagnet. The magnon branches are degenerate. (b) Berry curvature of the α-branch. The Berry curvature for the β-branch is equal but opposite in sign. In both panels, the model parameters are $S = 5/2$, $J_1 = 1.54$ meV, $J_2 = -0.14$ meV, $J_3 = 0.3$ meV, $J_z = 8.6$ μeV and $B = 0$, as appropriate for $MnPS_3$ [2, 3].

The Berry curvature of the magnon bands is defined as $\Omega_i(\mathbf{k}) = -\operatorname{Im}\langle \nabla_{\mathbf{k}} \Psi_i | \times \tau_z | \nabla_{\mathbf{k}} \Psi_i \rangle$ [2]. For a two-band system, it may also be expressed in the form $\Omega(\mathbf{k}) = (2d^3)^{-1} \mathbf{d} \cdot (\partial_{k_x} \mathbf{d} \times \partial_{k_y} \mathbf{d})$, where $\mathbf{d} = (h_0, h_x, h_y)$. The magnon dispersion and the associated magnon Berry curvature of the α-branch are shown in Fig. 1, with exchange parameters taken from Refs. [2, 3].

We note that the magnon Hamiltonian is invariant under the combined symmetry $\mathcal{TI}$, where $\mathcal{T}$ is the time-reversal operator and $\mathcal{I}$ is a reflection in the inversion center located halfway along an $A - B$ bond. The $\mathcal{TI}$ symmetry implies that the magnon dispersion is even in $\mathbf{k}$. In the presence of DMIs the $\mathcal{TI}$ symmetry is broken, and the C_{6v} symmetry of the excitation spectrum reduces to that of the C_{3v} subgroup. The combined time-reversal and bond inversion symmetry requires $\Omega(\mathbf{k}) = -\Omega(-\mathbf{k})$. However, in contrast to ferromagnetic systems, inversion symmetry is broken already for a vanishing DMI. Therefore, a non-zero Berry curvature appears even for $D = 0$ and in fact $\Omega(\mathbf{k})$ is independent of D.

We now define the magnon spin current operator in the magnon basis [4–8]

$$\mathbf{J} = \frac{1}{V} \sum_{\mathbf{k}} \Phi_{\mathbf{k}}^\dagger \begin{pmatrix} \mathbf{v}_{\alpha\mathbf{k}} & \mathbf{K}_{\mathbf{k}} \\ \mathbf{K}_{\mathbf{k}}^* & \mathbf{v}_{\beta\mathbf{k}} \end{pmatrix} \Phi_{\mathbf{k}}, \quad (5)$$

where $\Phi_{\mathbf{k}}^\dagger = (\alpha_{\mathbf{k}}^\dagger, \beta_{-\mathbf{k}})$, $\mathbf{v}_{i\mathbf{k}} = \nabla_{\mathbf{k}} \epsilon_{i\mathbf{k}}$ are the magnon velocities, and $\mathbf{K}_{\mathbf{k}} = -e^{-i\phi_{\mathbf{k}}}[(h_0 \nabla_{\mathbf{k}} d_0 - d_0 \nabla_{\mathbf{k}} h_0)/d - i(h_x \nabla_{\mathbf{k}} h_y - h_y \nabla_{\mathbf{k}} h_x)/d_0]$ with $d_0 = (h_x^2 + h_y^2)^{1/2}$. We note that for $D = 0$ the magnon velocity $\mathbf{v}_{-\mathbf{k}} = -\mathbf{v}_{\mathbf{k}}$ is odd since the magnon dispersion is even. The off-diagonal elements $\mathbf{K}_{\mathbf{k}}$ are related to the Berry curvature via $\Omega(\mathbf{k}) = (i/4d_{\mathbf{k}}^2) \mathbf{K}_{\mathbf{k}}^* \times \mathbf{K}_{\mathbf{k}}$, which is strongly reminiscent of the relation between the Berry curvature and interband matrix elements in electronic systems [9].

The optomagnetic Raman interaction Hamiltonian which describes the two-magnon Raman scattering in first order perturbation theory is given by

$$H_R = \sum_{ijq'q} R_{\mathbf{q}\mathbf{q}'} (\hat{\mathbf{e}}_q^* \cdot \boldsymbol{\delta}_j^{(1)})(\hat{\mathbf{e}}_{q'} \cdot \boldsymbol{\delta}_j^{(1)}) a_q^\dagger a_{q'} \mathbf{S}_i \cdot \mathbf{S}_{i+\delta_j^{(1)}}, \quad (6)$$

where the sum over j is over nearest neighbors. Transforming to Fourier space and rotating to the magnon basis, we find

$$H_R = S \sum_{\mathbf{k}q'q} R_{\mathbf{q}\mathbf{q}'} \Phi_{\mathbf{k}}^\dagger \begin{pmatrix} r_{\mathbf{k}q'q} & t_{\mathbf{k}q'q} \\ t_{\mathbf{k}q'q}^* & r_{\mathbf{k}q'q} \end{pmatrix} \Phi_{\mathbf{k}} a_{q'}^\dagger a_q. \quad (7)$$

Here $R_{\mathbf{q}\mathbf{q}'} = J_1(ea/\hbar)^2 \gamma_{\mathbf{q}} \gamma_{\mathbf{q}'}$ and the matrix elements are given by

$$\begin{aligned} r_{\mathbf{k}q'q} &= g_{0q'q} \cosh\theta_{\mathbf{k}} + \frac{1}{2}\sinh\theta_{\mathbf{k}} \big(g_{\mathbf{k}q'q} e^{i\phi_{\mathbf{k}}} + g_{\mathbf{k}q'q}^* e^{-i\phi_{\mathbf{k}}}\big) \\ t_{\mathbf{k}q'q} &= g_{0q'q} \sinh\theta_{\mathbf{k}} \\ &\quad + g_{\mathbf{k}q'q} e^{i\phi_{\mathbf{k}}} \cosh^2\frac{\theta_{\mathbf{k}}}{2} + g_{\mathbf{k}q'q}^* e^{-i\phi_{\mathbf{k}}} \sinh^2\frac{\theta_{\mathbf{k}}}{2} \end{aligned} \quad (8)$$

The light-matter interaction is controlled by the function $g_{\mathbf{k}q'q} = \sum_i (\hat{\mathbf{e}}_{q'}^* \cdot \boldsymbol{\delta}_i^{(1)})(\hat{\mathbf{e}}_q \cdot \boldsymbol{\delta}_i^{(1)}) e^{-i\mathbf{k}\cdot\boldsymbol{\delta}_i^{(1)}}$.

GENERAL FORM OF THE PHOTO-INDUCED MAGNON CURRENT

We here present the general form of the photo-induced magnon current arising from a symmetry analysis of the susceptibility tensor. As discussed in the main text, we define the optical susceptibility by

$$\langle J_{i_1} \rangle = \sigma_{i_1 i_2 i_3 i_4 i_5}(\omega) e_{in,i_2} e_{in,i_3}^* e_{sc,i_4}^* e_{sc,i_5}. \quad (9)$$

For a system invariant under the symmetry operation T_{ij}, the transformed suscptibility

$$\sigma'_{i_1 i_2 i_3 i_4 i_5} = \sum_{\substack{j_1 j_2 j_3 \\ j_4 j_5}} T_{i_1 j_1} T_{i_2 j_2} T_{i_3 j_3} T_{i_4 j_4} T_{i_5 j_5} \sigma_{j_1 j_2 j_3 j_4 j_5} \quad (10)$$

must equal the original susceptibility σ. As the present system is invariant under the symmetry group C_{3v}, the symmetry operations T are rotations around the z-axis by an angle $2\pi n/3$ for $n \in \{0,1,2\}$, as well as reflections in the planes through the origin with normal vectors $\hat{\mathbf{n}} = \hat{\mathbf{e}}_x$, $\hat{\mathbf{n}} = -\frac{1}{2}\hat{\mathbf{e}}_x + \frac{\sqrt{3}}{2}\hat{\mathbf{e}}_y$ and $\hat{\mathbf{n}} = -\frac{1}{2}\hat{\mathbf{e}}_x - \frac{\sqrt{3}}{2}\hat{\mathbf{e}}_y$. The first reflection, taking $x \to -x$, eliminates all elements with an odd number of indexes equal to x thus reducing the number of non-zero elements from 32 to 16. Imposing the remaining symmetries leaves five of these elements independent, which we take as σ_{yxyyx}, σ_{yyxxy}, σ_{yyxyx}, σ_{yyyxx} and σ_{yyyyy}. Due to the permutation symmetries $i_2 \leftrightarrow i_4$, $i_3 \leftrightarrow i_5$, the first, second and fourth of these elements are identical, leaving the three independent elements $\sigma_1 = \sigma_{yyxxy}$, $\sigma_2 = \sigma_{yyyyy}$ and $\sigma_3 = \sigma_{yyxyx}$.

The propagation direction of the incident field is determined by the unit vector $\hat{\mathbf{n}}$, which makes an angle θ with the z-axis and an angle ϕ with the x-axis. A right-handed coordinate system is defined by

$$\begin{aligned} \hat{\mathbf{e}}_1 &= -\sin\phi\hat{\mathbf{e}}_x + \cos\phi\hat{\mathbf{e}}_y \\ \hat{\mathbf{e}}_2 &= \cos\phi\cos\theta\hat{\mathbf{e}}_x + \sin\phi\cos\theta\hat{\mathbf{e}}_y + \sin\theta\hat{\mathbf{e}}_z \\ \hat{\mathbf{n}} &= \cos\phi\sin\theta\hat{\mathbf{e}}_x + \sin\phi\sin\theta\hat{\mathbf{e}}_y - \cos\theta\hat{\mathbf{e}}_z, \end{aligned} \quad (11)$$

which satisfy $\hat{\mathbf{e}}_1 \cdot \hat{\mathbf{e}}_2 = 0$, $\hat{\mathbf{e}}_i \cdot \hat{\mathbf{n}} = 0$ and $\hat{\mathbf{e}}_1 \times \hat{\mathbf{e}}_2 = \hat{\mathbf{n}}$. Assuming the direction of the incoming and scattered fields are identical, as imposed by the dipole approximation, we therefore have $\mathbf{n}_{sc} = \mathbf{n}_{in}$ and

$$\begin{aligned} \mathbf{n}_{in} &= \cos\phi\sin\theta\hat{\mathbf{e}}_x + \sin\phi\sin\theta\hat{\mathbf{e}}_y - \cos\theta\hat{\mathbf{e}}_z \\ \hat{\mathbf{e}}_{in} &= \frac{1}{\sqrt{2}}(\hat{\mathbf{e}}_1 - i\zeta\hat{\mathbf{e}}_2) \\ &= -\frac{1}{\sqrt{2}}(\sin\phi + i\zeta\cos\phi\cos\theta)\hat{\mathbf{e}}_x \\ &\quad + \frac{1}{\sqrt{2}}(\cos\phi - i\zeta\sin\phi\cos\theta)\hat{\mathbf{e}}_y, \end{aligned} \quad (12)$$

where for the polarization we only specify the x- and y-coordinates since only these couple to the system.

Using these expressions for the polarization, the relations among the susceptibility elements induced by the C_{3v} symmetry, and summing over the independent polarization directions of the scattered photons, the photo-induced magnon current in the x- and y-direction is given by

$$\begin{aligned} \langle \mathbf{J}_x \rangle &= \frac{1}{2}(\sigma_1 + \sigma_2)(\cos 2\theta + 3)\sin 2\phi \sin^2\theta \\ &\quad + \sigma_1 \sin 4\phi \sin^4\theta - i\zeta(\sigma_1 - \sigma_3)\cos\theta\cos 2\phi\sin^2\theta \\ \langle \mathbf{J}_y \rangle &= \frac{1}{2}(\sigma_1 + \sigma_2)(\cos 2\theta + 3)\cos 2\phi \sin^2\theta \\ &\quad - \sigma_1 \cos 4\phi \sin^4\theta + i\zeta(\sigma_1 - \sigma_3)\cos\theta\sin 2\phi\sin^2\theta. \end{aligned} \quad (13)$$

Only the tensor element σ_3 corresponds to a process with net spin angular momentum transfer and can contribute to the magnon current. Together with the requirement of a real current, this gives Eq. (1) of the main text

$$\langle \mathbf{J} \rangle = \zeta\,\mathrm{Im}(\sigma)\cos\theta\sin^2\theta(\sin 2\phi\hat{\mathbf{e}}_y - \cos 2\phi\hat{\mathbf{e}}_x). \quad (14)$$

DERIVATION OF THE OPTICAL SUSCEPTIBILITY

We consider the optical response to an external perturbation by writing the total Hamiltonian as $H = H_0 + H_R$, where H_R is assumed to be small. Here the equilibrium Hamiltonian is $H_0 = H_s + \sum_{\mathbf{k}} \hbar\omega_{\mathbf{k}} a^\dagger_{\mathbf{k}} a_{\mathbf{k}}$, where H_s is the spin Hamiltonian of Eq.2Main-text written in the Bogoliubov basis, and $a^\dagger_{\mathbf{k}}$ creates a photon in mode $\mathbf{k}$ of energy $\hbar\omega_{\mathbf{k}}$. Similarly H_R is the Raman interaction Hamiltonian of Eq.3Main-text.

The second order response of an observable J_μ is obtained by expanding the time evolution operators in the expectation value $\langle J_\mu\rangle(t) = \langle U(t_0,t) J_\mu U(t,t_0)\rangle$ to second order in H_R, and doing this we find

$$\begin{aligned}\langle J_\mu\rangle(t) &= \langle J_\mu\rangle_0(t) + i\int_{t_0}^{t} dt_1\, \langle [J_\mu(t), H_R(t_1)]\rangle_0 \\ &- \int_{t_0}^{t}\int_{t_0}^{t_1} dt_1 dt_2\, \langle [[J_\mu(t), H_R(t_1)], H_R(t_2)]\rangle_0\end{aligned} \tag{15}$$

The expectation value $\langle\cdots\rangle_0$ indicates a trace with the equilibrium density matrix, and all operators are in the Heisenberg picture with respect to the Hamiltonian H_0.

To evaluate the second order term we insert complete sets of eigenstates of H_0 between all operators, and introducing a phenomenological damping Γ_i to regularize the integral over t_i. Taking the Fourier transform of $\langle J\rangle$ we find only the static component is non-zero, and permuting some of the indexes gives the general expression

$$\begin{aligned}&\langle J\rangle \\ &= \sum_{mnl}\left[\frac{J_{ml}H_R^{ln}}{\epsilon_l - \epsilon_m - i\Gamma_1} - \frac{H_R^{ml}J_{ln}}{\epsilon_n - \epsilon_l - i\Gamma_1}\right]\frac{\rho_m H_R^{nm}}{\epsilon_n - \epsilon_m - i\Gamma_2} \\ &- \sum_{mnl}\frac{\rho_n H_R^{nm}}{\epsilon_n - \epsilon_m - i\Gamma_2}\left[\frac{J_{ml}H_R^{ln}}{\epsilon_l - \epsilon_m - i\Gamma_1} - \frac{H_R^{ml}J_{ln}}{\epsilon_n - \epsilon_l - i\Gamma_1}\right].\end{aligned} \tag{16}$$

To evaluate the matrix elements we assuming that the incoming field is initially in a number state $|n_{\mathbf{q}_{in}}\rangle_{s_{\mathbf{q}_{in}}}$ with polarization $s_{\mathbf{q}_{in}}$, and that the outgoing field is in the vacuum state $|0\rangle_s$ with polarization s. For the matrix elements to be non-zero, the right-most H_R in each line must then be proportional to $a^\dagger_q a_{q_{in}}$ and the left-most H_R to $a^\dagger_{q_{in}} a_q$. This means that the photon contribution to the energy difference $\epsilon_n - \epsilon_m$ is $-\Delta_{\mathbf{q}}$ in the first and $+\Delta_{\mathbf{q}}$ in the second term of Eq. 16.

Evaluating all possible combinations of the magnon operators giving a non-zero contribution, we find

$$\begin{aligned}\langle\mathbf{J}\rangle &= \frac{n_{\mathbf{q}_{in}}(RS)^2}{\pi V}\sum_{\mathbf{kq}s}\frac{n_{\mathbf{k}\alpha} + n_{\mathbf{k}\beta} + 1}{\hbar\omega_{\mathbf{q}_{in}}\hbar\omega_{\mathbf{q}}} \\ &\Bigg\{\left(\frac{1}{\epsilon_{\mathbf{k}} - \Delta_{\mathbf{q}} - i\Gamma_2} - \frac{e^{-\beta\epsilon_{\mathbf{k}}}}{\epsilon_{\mathbf{k}} + \Delta_{\mathbf{q}} - i\Gamma_2}\right)\times \\ &\left[\frac{t^*_{\mathbf{k}q_{in}q}(\mathbf{v}_{\alpha\mathbf{k}} + \mathbf{v}_{\beta\mathbf{k}})t_{\mathbf{k}qq_{in}}}{i\Gamma_1} + \frac{\mathbf{K}^*_{\mathbf{k}} r_{\mathbf{k}q_{in}q} t_{\mathbf{k}qq_{in}}}{\epsilon_{\mathbf{k}} - i\Gamma_1}\right] \\ &+\left(\frac{1}{\epsilon_{\mathbf{k}} - \Delta_{\mathbf{q}} + i\Gamma_2} - \frac{e^{-\beta\epsilon_{\mathbf{k}}}}{\epsilon_{\mathbf{k}} + \Delta_{\mathbf{q}} + i\Gamma_2}\right)\times \\ &\left[\frac{t^*_{\mathbf{k}q_{in}q}(\mathbf{v}_{\alpha\mathbf{k}} + \mathbf{v}_{\beta\mathbf{k}})t_{\mathbf{k}qq_{in}}}{-i\Gamma_1} + \frac{t^*_{\mathbf{k}q_{in}q} r_{\mathbf{k}qq_{in}} \mathbf{K}_{\mathbf{k}}}{\epsilon_{\mathbf{k}} - i\Gamma_1}\right]\Bigg\}.\end{aligned} \tag{17}$$

From this expression we can identify the Stokes component as the first terms in the parentheses. Here, the temperature effects come from the the factor $(n_{\mathbf{k}\alpha} + n_{\mathbf{k}\beta} + 1)$, which describes the thermal occupation of the magnon pair states. In the zero temperature limit, this contribution reduces to $(n_{\mathbf{k}\alpha} + n_{\mathbf{k}\beta} + 1) \to 1$. Similarly, we can identify the anti-Stokes component as the second terms in the parentheses, which differs from the Stokes component by the factor $-e^{-\beta\epsilon_{\mathbf{k}}}$. However, since the magnon spectrum is gapped, this term vanishes for $T \to 0$, so that only the Stokes component survives in the zero temperature limit.

Finally, taking the limit $\Gamma_i \to 0$ and evaluating the sum over $\mathbf{q}$, and we find the Stokes and anti-Stokes currents

$$\begin{aligned}&\langle\mathbf{J}\rangle_{st}(\mathbf{q}_{in}, s_{in}) = 2G\sum_s\int\frac{d\mathbf{k}^2}{(2\pi)^2}\,(n_{\mathbf{k}\alpha} + n_{\mathbf{k}\beta} + 1) \\ &\Bigg(\frac{t^*_{\mathbf{k}q_{in}q}(\mathbf{v}_{\alpha\mathbf{k}} + \mathbf{v}_{\beta\mathbf{k}})t_{\mathbf{k}qq_{in}}}{\Gamma\hbar\omega_{\mathbf{q}_{in}}}\Delta_{\mathbf{k}} \\ &+\frac{\mathbf{K}^*_{\mathbf{k}} r_{\mathbf{k}q_{in}q} t_{\mathbf{k}qq_{in}} + t^*_{\mathbf{k}q_{in}q} r_{\mathbf{k}qq_{in}}\mathbf{K}_{\mathbf{k}}}{\pi\hbar\omega_{\mathbf{q}_{in}}\epsilon_{\mathbf{k}}}\left[\hbar\omega_{\mathbf{q}_{in}} + \Delta_{\mathbf{k}}\ln\frac{\epsilon_{\mathbf{k}}}{\Delta_{\mathbf{k}}}\right] \\ &+ i\frac{\mathbf{K}^*_{\mathbf{k}} r_{\mathbf{k}q_{in}q} t_{\mathbf{k}qq_{in}} - t^*_{\mathbf{k}q_{in}q} r_{\mathbf{k}qq_{in}}\mathbf{K}_{\mathbf{k}}}{\hbar\omega_{\mathbf{q}_{in}}\epsilon_{\mathbf{k}}}\Delta_{\mathbf{k}}\Bigg)\end{aligned} \tag{18}$$

$$\begin{aligned}&\langle\mathbf{J}\rangle_{as}(\mathbf{q}_{in}, s_{in}) = -2G\sum_s\int\frac{d\mathbf{k}^2}{(2\pi)^2}\,(n_{\mathbf{k}\alpha} + n_{\mathbf{k}\beta} + 1)e^{-\beta\epsilon_{\mathbf{k}}} \\ &\Bigg(\frac{t^*_{\mathbf{k}q_{in}q}(\mathbf{v}_{\alpha\mathbf{k}} + \mathbf{v}_{\beta\mathbf{k}})t_{\mathbf{k}qq_{in}}}{\Gamma\hbar\omega_{\mathbf{q}_{in}}}\bar{\Delta}_{\mathbf{k}} \\ &-\frac{\mathbf{K}^*_{\mathbf{k}} r_{\mathbf{k}q_{in}q} t_{\mathbf{k}qq_{in}} + t^*_{\mathbf{k}q_{in}q} r_{\mathbf{k}qq_{in}}\mathbf{K}_{\mathbf{k}}}{\pi\hbar\omega_{\mathbf{q}_{in}}\epsilon_{\mathbf{k}}}\left[\hbar\omega_{\mathbf{q}_{in}} + \bar{\Delta}_{\mathbf{k}}\ln\frac{\epsilon_{\mathbf{k}}}{\bar{\Delta}_{\mathbf{k}}}\right] \\ &+ i\frac{\mathbf{K}^*_{\mathbf{k}} r_{\mathbf{k}q_{in}q} t_{\mathbf{k}qq_{in}} - t^*_{\mathbf{k}q_{in}q} r_{\mathbf{k}qq_{in}}\mathbf{K}_{\mathbf{k}}}{\hbar\omega_{\mathbf{q}_{in}}\epsilon_{\mathbf{k}}}\bar{\Delta}_{\mathbf{k}}\Bigg).\end{aligned} \tag{19}$$

Here, top simplify the notation, we have written $\epsilon_{\mathbf{k}} = \epsilon_{\alpha\mathbf{k}} + \epsilon_{\beta\mathbf{k}}$, $\Delta_{\mathbf{k}} = \hbar\omega_{\mathbf{q}_{in}} - \epsilon_{\mathbf{k}}$ and $\bar{\Delta}_{\mathbf{k}} = \hbar\omega_{\mathbf{q}_{in}} + \epsilon_{\mathbf{k}}$. Also, Γ^{-1} is the magnon lifetime, $\beta = (k_B T)^{-1}$ and $G = n_{\mathbf{q}_{in}}(RS)^2 A/(2\pi\hbar c)^3$. Using the relation $n_{\mathbf{q}_{in}} = IV/(\hbar\omega_{\mathbf{q}_{in}} c)$, we find

$$G = \left(\frac{I}{\hbar\omega_{\mathbf{q}_{in}} c}\right)\left(\frac{A}{V}\right)\left(\frac{J_1^2 S^2 e^4 a^4}{32\pi^3\hbar^3 c^3\epsilon_0^2}\right). \tag{20}$$

Assuming $I = 10^{12}$ Wcm^{-2}, $\hbar\omega_{\mathbf{q}_{in}} = 1$ eV and a vertical extent $z = 5$ Å of the system, we have

$$G = 5.78914\cdot 10^{-24}\ \mathrm{Jm}^{-1} \tag{21}$$

RELATION BETWEEN INVERSE SPIN HALL VOLTAGE AND MAGNON CURRENT

Following the discussion in Ref. [10], we first consider the conversion of the magnon current to a spin current at the $MnPS_3$/Pt interface. Using a Boltzmann equation to model the magnon diffusion and accumulation at the interface, the authors of Ref. [10] find the spin current

$$j_s = \frac{\lambda_N}{d_N}\frac{b(\cosh[d_N/\lambda_N]-1)}{(1+b)\sinh[d_N/\lambda_N]}\langle \mathbf{J}\rangle_\perp, \tag{22}$$

where λ_N is the spin diffusion length of Pt, d_N the thickness of the Pt layer and $b = G_{me}l_m/D_m$. Further, l_m is the magnon diffusion length, $D_m = l_m^2/\tau_m$ the diffusion coefficient, and τ_m the magnon non-conserving lifetime of $MnPS_3$. Finally G_{me} is the conversion coefficient from a magnon to spin current, given by

$$G_{me} = (\pi S/\hbar)J_{sd}^2 g_e(\epsilon_F)^2 a_{\mathrm{Pt}}^2 a_{\mathrm{MnPS_3}}^5 \bar{E}_m, \tag{23}$$

where J_{sd} is the electron-magnon exchange coupling, g_e the Pt density of states at the Fermi energy, a_{Pt} and $a_{\mathrm{MnPS_3}}$ the lattice constants of Pt and $MnPS_3$ respectively, and $\bar{E}_m$ the mean magnon lifetime. Using the parameter values $\lambda_N = d_N = 1$ nm, $l_m = 3$ μm [11] and $\tau_m = 10$ μs, as well as $J_{sd} = 1$ meV, $g_e = 0.08$ $(\mathrm{eVa_{Pt}^3})^{-1}$, $a_{\mathrm{Pt}} = 3.9$ Å, $a_{\mathrm{MnPS_3}} = 6.1$ Å and $\bar{E}_m = 5$ meV, we find $G_{me} = 0.37$. This gives a conversion factor between j_s and $\langle \mathbf{J}\rangle_\perp$ of $C_{sm} = 0.054$.

We now consider the conversion of the induced spin current to a voltage V_{ISHE} via the inverse spin Hall effect (ISHE). Following Ref. [12], we find the ISHE voltage is given by

$$V_{\mathrm{ISHE}} = \frac{2e}{\hbar}\frac{w\theta_{\mathrm{SHE}}\lambda_N \tanh(d_N/2\lambda_N)}{d_N\sigma_N} j_s, \tag{24}$$

where w is the width of the $MnPS_3$ layer, and θ_{SHE} and σ_N are the spin Hall angle and the electrical conductivity of Pt. Using the parameters values $w = 1$ mm, $\theta_{\mathrm{SHE}} = 0.04$ and $\sigma_N = 2$ $(\mu\Omega\mathrm{m})^{-1}$, we find a conversion factor between V_{ISHE} and j_s of $C_{Vs} = 28000$. Thus, the ISHE voltage measured in response to the photo-generated magnon current is given by

$$V_{\mathrm{ISHE}} = C_{Vs}C_{sm}\langle \mathbf{J}\rangle_\perp \approx (170\mathrm{Vm^2/J})\langle \mathbf{J}\rangle_\perp, \tag{25}$$

for a magnon current given in $\mathrm{J/m^2}$.

This results assumes the magnon current is generated right at the $MnPS_3$/Pt interface. In practice, there would be an attenuation of the magnon current associated with the propagation through the magnetic substrate. Although the attenuation is hard to predict, a rough estimate can be obtained by comparing the magnon diffusion length l_m to the thickness d_m of the $MnPS_3$ layer and assuming an exponential decay. Thus, we use $\alpha = e^{-d_m/l_m}$ as our attenuation factor. Assuming $d_m = 10$ μm we find $\alpha = 0.04$, and a voltage V_{ISHE} on the order of mV.

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