Supplemental Material for "Observing Altermagnetism using Polarized Neutrons"

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REVIEW OF POLARIZED NEUTRON SCATTERING CROSS SECTIONS

This section contains a short self-contained review of neutron cross sections from magnetic crystals when either the beam is polarized or the detectors are sensitive to polarization. See also Refs. [1, 2] for a more comprehensive treatment.

The in-going neutron beam will be assumed to have polarization $\boldsymbol{P}_{\rm in} \equiv 2\langle \boldsymbol{I}\rangle$ where $\boldsymbol{I}$ is the neutron spin. Therefore $0 \leq |\boldsymbol{P}_{\rm in}| \leq 1$ where $|\boldsymbol{P}_{\rm in}| = 0$ describes an unpolarized beam and $|\boldsymbol{P}_{\rm in}| = 1$ is a fully polarized beam along $\hat{\boldsymbol{P}}_{\rm in}$. The polarization of scattered neutrons is denoted as $\boldsymbol{P}_{\rm out}$.

The ions in the crystal are indexed by $\boldsymbol{r}_n = \boldsymbol{r} + \boldsymbol{\delta}_n$ where $\boldsymbol{r}$ are the primitive lattice vectors of the magnetic unit cell and $\boldsymbol{\delta}_n$ runs over ions in the this unit cell. Each atom carries a moment $\boldsymbol{\mu}_{\boldsymbol{r},n} = g_n\mu_{\rm B}\boldsymbol{S}_{\boldsymbol{r},n}$ (that are zero for the non-magnetic ions). We introduce b_n for the coherent scattering length of the ions.

For elastic scattering of neutrons the cross section can be broken into three parts

$$\frac{d\sigma}{d\Omega} = \left(\frac{d\sigma}{d\Omega}\right)_{\rm nuc} + \left(\frac{d\sigma}{d\Omega}\right)_{\rm mag} + \left(\frac{d\sigma}{d\Omega}\right)_{\rm nuc-mag}, \tag{S1}$$

the cross section from nuclear scattering of unpolarized neutrons, the part coming from scattering of unpolarized neutrons from magnetic moments and the nuclear-magnetic interference term. The last term can be written

$$\left(\frac{d\sigma}{d\Omega}\right)_{\rm nuc-mag} \propto \sum_{\boldsymbol{r},\boldsymbol{r}'} e^{i\boldsymbol{k}\cdot(\boldsymbol{r}-\boldsymbol{r}')}\boldsymbol{P}^{\perp}_{\rm in}\cdot {\rm Re}\left[\boldsymbol{F}^{*}_{\rm mag}(\boldsymbol{k})F_{\rm nuc}(\boldsymbol{k})\right] \tag{S2}$$

where we have define the form factors

$$F_{\rm nuc}(\boldsymbol{k}) \equiv \sum_n e^{i\boldsymbol{k}\cdot\boldsymbol{\delta}_n} b_n \tag{S3}$$

$$\boldsymbol{F}_{\rm mag}(\boldsymbol{k}) \equiv \mu_{\rm B}\sum_n e^{i\boldsymbol{k}\cdot\boldsymbol{\delta}_n} g_n f_n(\boldsymbol{k})\langle \boldsymbol{S}_n\rangle = \langle \boldsymbol{M}_{\boldsymbol{k}}\rangle, \tag{S4}$$

where the static moment does not depend on the unit cell $\langle \boldsymbol{S}_{\boldsymbol{r},n}\rangle \equiv \langle \boldsymbol{S}_n\rangle$. We have adopted the notation $\boldsymbol{P}^{\perp}_{\rm in} \equiv \hat{\boldsymbol{k}}\times(\boldsymbol{P}_{\rm in}\times\hat{\boldsymbol{k}})$ for any vector quantity that appears below.

This reduces to Eq. (1) of the main text when when the moments are colinear. This contribution vanishes when the scattering wavevector is aligned with the in-going polarization direction or when the moments are colinear and aligned with $\boldsymbol{k}$. Of particular interest to us is the fact that the contribution of Eq. (S2), out of the entire elastic cross section, is time reversal odd so it can potentially distinguish domains related by time reversal symmetry.

The purely magnetic scattering cross section can also be split into the usual unpolarized part and a polarized part

$$\left(\frac{d\sigma}{d\Omega}\right)_{\rm mag} = \left(\frac{d\sigma}{d\Omega}\right)_{\rm mag,unpol} + \left(\frac{d\sigma}{d\Omega}\right)_{\rm mag,pol}.$$

The purely magnetic polarized contribution takes the form

$$\left(\frac{d\sigma}{d\Omega}\right)_{\rm mag,pol} \propto -i\sum_{\boldsymbol{r},\boldsymbol{r}'} e^{i\boldsymbol{k}\cdot(\boldsymbol{r}-\boldsymbol{r}')}\boldsymbol{P}_{\rm in}\cdot\left[\boldsymbol{F}^{\perp}_{\rm mag}(\boldsymbol{k})\right]^{*}\times\boldsymbol{F}^{\perp}_{\rm mag}(\boldsymbol{k}). \tag{S5}$$

Since this part is time reversal invariant it does not contain information about the domain populations.

The cross section for inelastic scattering of neutrons can be decomposed in an similar way. Here we consider only the purely magnetic contributions, starting with the usual unpolarized case

$$\left(\frac{d^2\sigma}{d\Omega d\omega}\right)_{\rm mag,unpol} \propto \int_{-\infty}^{+\infty} dt e^{-i\omega t}\langle \boldsymbol{M}^{\perp}_{-\boldsymbol{k}}(0)\cdot\boldsymbol{M}^{\perp}_{\boldsymbol{k}}(t)\rangle, \tag{S6}$$

and the polarization dependent contribution

$$\left(\frac{d^2\sigma}{d\Omega d\omega}\right)_{\rm mag,pol} \propto +i\int_{-\infty}^{+\infty} dt e^{-i\omega t}\boldsymbol{P}_{\rm in}\cdot\langle \boldsymbol{M}^{\perp}_{-\boldsymbol{k}}(0)\times\boldsymbol{M}^{\perp}_{\boldsymbol{k}}(t)\rangle, \tag{S7}$$

which vanishes when the beam is unpolarized. Due to the cross products appearing in this expression we see that polarized neutrons evidently allow one to measure components of the antisymmetric spin-spin correlator.

Now we restrict our attention to the cross section for altermagnets in the zero spin-orbit coupling limit. The moments are assumed to be colinear along the $\hat{z}$ axis and we suppose that the Hamiltonian has a global $U(1)$ symmetry such that the magnons have a well-defined chirality. If we also put the beam polarization along the moment direction the cross section is

$$\frac{d^2\sigma}{d\Omega d\omega} \propto \frac{1}{2}\int_{-\infty}^{+\infty} dt e^{-i\omega t}\left[\left(\left\langle M^{+}_{-\boldsymbol{k}}(0)M^{-}_{\boldsymbol{k}}(t)\right\rangle + \left\langle M^{-}_{-\boldsymbol{k}}(0)M^{+}_{\boldsymbol{k}}(t)\right\rangle\right) + P^{z}_{\rm in}(\hat{k}_z)^2\left(\left\langle M^{+}_{-\boldsymbol{k}}(0)M^{-}_{\boldsymbol{k}}(t)\right\rangle - \left\langle M^{-}_{-\boldsymbol{k}}(0)M^{+}_{\boldsymbol{k}}(t)\right\rangle\right)\right]. \tag{S8}$$

The dynamical correlators $\left\langle M^{+}_{-\boldsymbol{k}}(0)M^{-}_{\boldsymbol{k}}(t)\right\rangle$ in this expression is non-vanishing for one chirality and vanishes for the other and *vice versa* for the other correlator.

Having considered the cross section for magnetic scattering of polarized neutrons we now examine expressions for the polarization of the scattered beam. This polarization is written as the ratio of a correlator with intensity for the unpolarized case

$$\boldsymbol{P}_{\rm out} = \left(\frac{d^2\sigma}{d\Omega d\omega}\right)^{-1}_{\boldsymbol{P}_{\rm in}=0} \int_{-\infty}^{+\infty} dt e^{-i\omega t} \left[\left\langle \boldsymbol{M}^{\perp}_{-\boldsymbol{k}}(0)\left(\boldsymbol{P}_{\rm in}\cdot \boldsymbol{M}^{\perp}_{\boldsymbol{k}}(t)\right)\right\rangle + \left\langle\left(\boldsymbol{P}_{\rm in}\cdot \boldsymbol{M}^{\perp}_{-\boldsymbol{k}}(0)\right)\boldsymbol{M}^{\perp}_{\boldsymbol{k}}(t)\right\rangle - \boldsymbol{P}_{\rm in}\left\langle \boldsymbol{M}^{\perp}_{-\boldsymbol{k}}(0)\cdot \boldsymbol{M}^{\perp}_{\boldsymbol{k}}(t)\right\rangle - i\left\langle \boldsymbol{M}^{\perp}_{-\boldsymbol{k}}(0)\times \boldsymbol{M}^{\perp}_{\boldsymbol{k}}(t)\right\rangle\right].$$

Restricting our attention to altermagnetism again, suppose the moments are aligned along $\hat{z}$ and have a $U(1)$ symmetry and for simplicity that the ingoing beam is unpolarized. Then the outgoing polarization is given by

$$\boldsymbol{P}_{\rm out} = +\frac{1}{2}\hat{\boldsymbol{k}}\hat{k}_z \left(\frac{d^2\sigma}{d\Omega d\omega}\right)^{-1}_{P_{\rm in}=0} \int_{-\infty}^{+\infty} dt e^{-i\omega t}\left[\left\langle M^{-}_{-\boldsymbol{k}}(0) M^{+}_{\boldsymbol{k}}(t)\right\rangle - \left\langle M^{+}_{-\boldsymbol{k}}(0) M^{-}_{\boldsymbol{k}}(t)\right\rangle\right]. \tag{S9}$$

We thus see that the outgoing polarization is directly related to the magnon chirality.

REVIEW OF LINEAR SPIN-WAVE THEORY

Here we briefly review linear spin-wave theory to establish notation used in the main text. We consider a spin Hamiltonian

$$H = \sum_{\boldsymbol{r}\boldsymbol{\delta}}\sum_{nn'} \boldsymbol{S}^{\intercal}_{\boldsymbol{r},n}\boldsymbol{J}_{\boldsymbol{\delta},nn'}\boldsymbol{S}_{\boldsymbol{r}+\boldsymbol{\delta},n'}$$

where $\boldsymbol{r}, n$ denotes the unit cell and sublattice index of the spin. We now consider a semi-classical expansion about some magnetically ordered ground state. The spin operators can expressed in terms of Holstein-Primakoff bosons as

$$\boldsymbol{S}_{\boldsymbol{r},n} \equiv \sqrt{S}\left[\left(1-\frac{a^{\dagger}_{\boldsymbol{r},n}a_{\boldsymbol{r},n}}{2S}\right)^{1/2} a_{\boldsymbol{r},n}\hat{\boldsymbol{e}}_{n,-} + a^{\dagger}_{\boldsymbol{r},n}\left(1-\frac{a^{\dagger}_{\boldsymbol{r},n}a_{\boldsymbol{r},n}}{2S}\right)^{1/2}\hat{\boldsymbol{e}}_{n,+}\right] + \left(S - a^{\dagger}_{\boldsymbol{r},n}a_{\boldsymbol{r},n}\right)\hat{\boldsymbol{e}}_{n,0},$$

where the vectors $\hat{\boldsymbol{e}}_{n,\pm}$, $\hat{\boldsymbol{e}}_{n,0}$ define a local frame of reference; in a more conventional Cartesian basis one defines $\hat{\boldsymbol{e}}_{n,\pm} \equiv (\hat{\boldsymbol{x}}_n \pm i\hat{\boldsymbol{y}}_n)/\sqrt{2}$ and $\hat{\boldsymbol{e}}_{n,0} \equiv \hat{z}_n$. It is useful to write the exchange matrix in the frame aligned with these axes

$$\mathcal{J}^{\mu\mu'}_{\boldsymbol{\delta},nn'} \equiv \hat{\boldsymbol{e}}^{\intercal}_{n,\mu}\boldsymbol{J}_{\boldsymbol{\delta},nn'}\hat{\boldsymbol{e}}_{n',\mu'}, \tag{S10}$$

with $\mu = 0, \pm$. Expanding in powers of $1/S$ then yields a semi-classical expansion about the ordered state defined by $\hat{\boldsymbol{e}}_{n,0}$.

At order $O(S)$ in the Holstein-Primakoff operators we have

$$\boldsymbol{S}_{\boldsymbol{r},n} \approx \sqrt{S}\left[a_{\boldsymbol{r},n}\hat{\boldsymbol{e}}_{n,-} + a^{\dagger}_{\boldsymbol{r},n}\hat{\boldsymbol{e}}_{n,+}\right] + \left(S - a^{\dagger}_{\boldsymbol{r},n}a_{\boldsymbol{r},n}\right)\hat{\boldsymbol{e}}_{n,0}, \tag{S11}$$

Inserting this into our spin Hamiltonian and keeping only terms to $O(S)$ yields

$$H = E_0 + \frac{1}{2}\sum_{\boldsymbol{k}}\left([\boldsymbol{a}^{\dagger}_{\boldsymbol{k}}]^{\intercal}\ \boldsymbol{a}^{\intercal}_{-\boldsymbol{k}}\right)\begin{pmatrix}\boldsymbol{A}_{\boldsymbol{k}} & \boldsymbol{B}_{\boldsymbol{k}}\\ \boldsymbol{B}^{*}_{-\boldsymbol{k}} & \boldsymbol{A}^{*}_{-\boldsymbol{k}}\end{pmatrix}\begin{pmatrix}\boldsymbol{a}_{\boldsymbol{k}}\\ \boldsymbol{a}^{\dagger}_{-\boldsymbol{k}}\end{pmatrix}, \tag{S12}$$

where E_0 is the classical energy, N_s is the number of sublattices, N is the total number of sites and we have defined the Fourier transforms of the bosons as $a_{\boldsymbol{k}n} \equiv N_c^{-1/2}\sum_{\boldsymbol{r}} e^{-i\boldsymbol{k}\cdot(\boldsymbol{r}+\boldsymbol{\delta}_n)}a_{\boldsymbol{r},n}$ where $N = N_cN_s$. The matrices $\boldsymbol{A}_{\boldsymbol{k}}$ and $\boldsymbol{B}_{\boldsymbol{k}}$ are given by

$$A^{nn'}_{\boldsymbol{k}} = S\left(\mathcal{J}^{+-}_{\boldsymbol{k},nn'} - \delta_{nn'}\sum_{n''}\mathcal{J}^{00}_{\boldsymbol{0},nn''}\right), \tag{S13a}$$

$$B^{nn'}_{\boldsymbol{k}} = S\mathcal{J}^{++}_{\boldsymbol{k},nn'}, \tag{S13b}$$

where we have defined the Fourier transforms of the local exchange matrices as

$$\mathcal{J}^{\mu\mu'}_{\boldsymbol{k},nn'} \equiv \sum_{\boldsymbol{d}}\mathcal{J}^{\mu\mu'}_{\boldsymbol{d},nn'}e^{i\boldsymbol{k}\cdot(\boldsymbol{d}+\boldsymbol{\delta}_{n'}-\boldsymbol{\delta}_n)}. \tag{S14}$$

The linear spin-wave Hamiltonian [Eq. (S12)] can be diagonalized by a Bogoliubov transformation. To do this one diagonalizes the modified matrix [3]

$$\begin{pmatrix}\boldsymbol{A}_{\boldsymbol{k}} & \boldsymbol{B}_{\boldsymbol{k}}\\ -\boldsymbol{B}^{*}_{-\boldsymbol{k}} & -\boldsymbol{A}^{*}_{-\boldsymbol{k}}\end{pmatrix} \equiv \boldsymbol{\sigma}_3\boldsymbol{M}_{\boldsymbol{k}} \tag{S15}$$

where $\boldsymbol{\sigma}_\mu$ denotes the set of block Pauli matrices. This yields pairs of eigenvectors $\boldsymbol{V}_{\boldsymbol{k},n}$ and $\boldsymbol{W}_{-\boldsymbol{k},n} = \boldsymbol{\sigma}_1\boldsymbol{V}^{*}_{-\boldsymbol{k},n}$ with eigenvalues $+\epsilon_{\boldsymbol{k},n}$ and $-\epsilon_{-\boldsymbol{k},n}$. These vectors can be normalized such that [3]

$$\boldsymbol{V}^{\dagger}_{\boldsymbol{k},n}\boldsymbol{\sigma}_3\boldsymbol{V}_{\boldsymbol{k},n'} = +\delta_{nn'}, \qquad \boldsymbol{W}^{\dagger}_{-\boldsymbol{k},n}\boldsymbol{\sigma}_3\boldsymbol{W}_{-\boldsymbol{k},n'} = -\delta_{nn'},$$
$$\boldsymbol{W}^{\dagger}_{-\boldsymbol{k},n}\boldsymbol{\sigma}_3\boldsymbol{V}_{\boldsymbol{k},n'} = 0.$$

One can then write the Hamiltonian in terms of diagonalized bosons, $\gamma_{\boldsymbol{k},n}$, as [3]

$$H = \sum_{\boldsymbol{k},n}\epsilon_{\boldsymbol{k},n}\gamma^{\dagger}_{\boldsymbol{k},n}\gamma_{\boldsymbol{k},n} + \text{const.} \tag{S16}$$

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