Supplementary Material for Strongly anisotropic spin dynamics in magnetic topological insulators

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I. DETAILS ON THE THEORY OF ESR FOR SPIN-ORBIT COUPLED SYSTEMS

Following the well-established theory of NMR, when the static field is applied along z-axis, the spin relaxation rate can be expressed as[1,2]

$$\frac{1}{\tau_z} = \frac{\gamma^2}{4}\int dt\, \cos\omega_L t \langle\{\delta B_+(t), \delta B_-(0)\}\rangle + \frac{\gamma^2}{2}\int dt\, \langle\{\delta B_z(t), \delta B_z(0)\}\rangle\ , \tag{1}$$

in terms of the fluctuating internal field $\delta\mathbf{B}$ acting on the local moment. Here γ denotes the gyromagnetic ratio, $\omega_L = \gamma H_{\rm res}$ is the resonance frequency, $\hat{A}, \hat{B} = \hat{A}\hat{B} + \hat{B}\hat{A}$ is the anticommutator of two operators, and finally $\langle\hat{A}\rangle = {\rm Tr}(\rho_{\rm th}\hat{A})$ denotes the statistical average of the operator. Also the field is decomposed as $\delta\mathbf{B} = \delta B_+\hat{\mathbf{e}}_- + \delta B_-\hat{\mathbf{e}}_+ + \delta B_z\hat{\mathbf{e}}_z$ with $\delta B_\pm = \delta B_x \pm i\delta B_y$ and $\hat{\mathbf{e}}_\pm = (\hat{\mathbf{e}}_x \pm i\hat{\mathbf{e}}_y)/2$. Notice that $\hat{\mathbf{e}}_\pm$ and $\hat{\mathbf{e}}_z$ also create an orthogonal coordinate frame.

We consider a bath of noninteracting conduction fermions $\mathcal{H}_0 = \sum_{\nu k}\varepsilon_{\nu\mathbf{k}}c^\dagger_{\nu\mathbf{k}}c_{\nu\mathbf{k}}$, where $\varepsilon_{\nu\mathbf{k}}$ are the energy bands of the corresponding Bloch wavefunctions $|\psi_{\nu\mathbf{k}}\rangle = e^{i\mathbf{k}\cdot\mathbf{r}}|u_{\nu\mathbf{k}}\rangle$. These fermions are coupled with a local moment $\mathbf{S}$ via a general anisotropic s-d exchange coupling, which reads

$$\mathcal{H}_{\rm ex} = \sum_{\mathbf{q}}\mathbf{S}\cdot\mathbf{J}\cdot\hat{s}_{\mathbf{q}} = \sum_{ij,\mathbf{q}}J_{ij}S^i\hat{s}^j_{\mathbf{q}},\ \text{with}\ \hat{s}^j_{\mathbf{q}} = \frac{1}{2}\sum_{\mathbf{k},s,s'}c^\dagger_{\mathbf{k}+\mathbf{q},s}\sigma^j_{ss'}c_{\mathbf{k},s'} \tag{2}$$

Here, we omit, for brevity, the band index. The fluctuating fields due to the electronic cloud around the local moments and in terms of electron creation and annihilation field operators, read

$$\widehat{\delta B}_\pm = \frac{1}{\gamma}\sum_{j,\mathbf{q}}J_{\pm,j}\hat{s}^j_{\mathbf{q}} = \frac{1}{\gamma}\sum_{j,\mathbf{q}}\left(J_{x,j} \pm iJ_{y,j}\right)\hat{s}^j_{\mathbf{q}} \tag{3}$$

$$\widehat{\delta B}_z = \frac{1}{\gamma}\sum_{j,\mathbf{q}}J_{z,j}\,\hat{s}^j_{\mathbf{q}} \tag{4}$$

Inserting above expressions in Eq. 1, we find:

$$\frac{1}{\tau_z} = \frac{1}{4}\sum_{i,j}J_{+,i}J_{-,j}\int dt\,\cos\omega_L t\sum_{\mathbf{q}}\langle\{\hat{s}^i_{\mathbf{q}}(t), \hat{s}^j_{-\mathbf{q}}\}\rangle + \frac{1}{2}\sum_{i,j}J_{z,i}J_{z,j}\int dt\sum_{\mathbf{q}}\langle\{\hat{s}^i_{\mathbf{q}}(t), \hat{s}^j_{-\mathbf{q}}\}\rangle \tag{5}$$

Using the symmetry of the anticommutator, $\langle\{\hat{s}^i_{\mathbf{q}}(t), \hat{s}^j_{-\mathbf{q}}\}\rangle = \langle\{\hat{s}^j_{-\mathbf{q}}(-t), \hat{s}^i_{\mathbf{q}}\}\rangle$, and noting that $\frac{J_{+,i}\,J_{-,j} + J_{-,i}\,J_{+,j}}{2} = J_{x,i}\,J_{x,j} + J_{y,i}\,J_{y,j}$ the relaxation time reads

$$\frac{1}{\tau_z} = T\sum_{i,j}\left(J_{x,i}\,J_{x,j} + J_{y,i}\,J_{y,j} + 2\,J_{z,i}\,J_{z,j}\right)\times\lim_{\omega\to 0}\frac{1}{\omega}\sum_{\mathbf{q}}{\rm Im}\,\chi_{ij}(iq_n, \mathbf{q})|_{iq_n\to\omega+i0^+} \tag{6}$$

To obtain this equation, we have used the quantum-mechanical version of fluctuation-dissipation theorem defining the spin susceptibility tensor $\chi(iq_n, \mathbf{q})$ with components $\chi_{ij}(iq_n, \mathbf{q})$. This relation is valid for large enough temperature $T \gg \omega_L$. Note that we have set the Planck and Boltzmann constant as $\hbar = k_B = 1$. The spin susceptibility in Matsubara representation is

$$\chi_{ij}(iq_n, \mathbf{q}) = -\frac{T}{4}\sum_{ik_n,\mathbf{k}}{\rm Tr}\left[\hat{\sigma}^i\,\hat{G}(ik_n, \mathbf{k})\,\hat{\sigma}^j\,\hat{G}(ik_n + iq_n, \mathbf{k}+\mathbf{q})\right], \tag{7}$$

with bosonic and fermionic Matsubara frequencies denoted as $q_n = 2n\pi T$ and $k_n = (2n+1)\pi T$, respectively. The noninteracting Green's function can be generally written as

$$\hat{G}(ik_n, \mathbf{k}) = \sum_{\nu} \frac{|u_{\nu\mathbf{k}}\rangle\langle u_{\nu\mathbf{k}}|}{ik_n - \varepsilon_{\nu\mathbf{k}}}. \tag{8}$$

We can readily arrive at the following suitable expression for the susceptibility

$$\chi_{ij}(iq_n, \mathbf{q}) = -\frac{T}{4} \sum_{ik_n,\mathbf{k}} \sum_{\nu\nu'} \mathrm{Tr}\big[\mathcal{F}^{i,\nu\nu'}_{\mathbf{k},\mathbf{k}+\mathbf{q}}\, \mathcal{F}^{j,\nu'\nu}_{\mathbf{k}+\mathbf{q},\mathbf{k}}\big] \times \frac{1}{(ik_n - \varepsilon_{\nu\mathbf{k}})(ik_n + iq_n - \varepsilon_{\nu',\mathbf{k}+\mathbf{q}})}, \tag{9}$$

with $\mathcal{F}^{i,\nu\nu'}_{\mathbf{k}\,\mathbf{k}'} = \langle u_{\nu\mathbf{k}}|\hat{\sigma}^i|u_{\nu'\mathbf{k}'}\rangle/2$. Then by performing the Matsubara sum we find

$$\frac{1}{\tau_z} = -\pi T \sum_{i,j} \big(J_{x,i}\, J_{x,j} + J_{y,i}\, J_{y,j} + 2\, J_{z,i}\, J_{z,j}\big) \sum_{\mathbf{k},\mathbf{q},\nu,\nu'} \delta(\varepsilon_{\nu\mathbf{k}} - \varepsilon_{\nu',\mathbf{k}+\mathbf{q}})\, \frac{\partial n(\omega)}{\partial \omega}\bigg|_{\omega=\varepsilon_{\nu\mathbf{k}}} \mathrm{Tr}\big[\mathcal{F}^{i,\nu\nu'}_{\mathbf{k},\mathbf{k}+\mathbf{q}}\, \mathcal{F}^{j,\nu'\nu}_{\mathbf{k}+\mathbf{q},\mathbf{k}}\big]. \tag{10}$$

Considering diagonal exchange coupling terms $J_{ij} = J_i\delta_{ij}$, defining $\mathrm{Tr}\big[\mathcal{F}^{i,\nu\nu'}_{\mathbf{k}\mathbf{k}'}\, \mathcal{F}^{i,\nu'\nu}_{\mathbf{k}'\mathbf{k}}\big] = |\mathcal{F}^{i,\nu\nu'}_{\mathbf{k}\mathbf{k}'}|^2$ and replacing the derivative of Fermi-Dirac distribution $n(\omega)$ with its zero temperature value, this simplifies to

$$\frac{1}{\tau_z} = \pi T \sum_{\nu\mathbf{k},\nu'\mathbf{k}'} \delta(\varepsilon_{\nu\mathbf{k}} - \varepsilon_{\nu'\mathbf{k}'})\, \delta(\varepsilon_F - \varepsilon_{\nu\mathbf{k}}) \times \Big(J_x^2 |\mathcal{F}^{x,\nu\nu'}_{\mathbf{k}\mathbf{k}'}|^2 + J_y^2 |\mathcal{F}^{y,\nu\nu'}_{\mathbf{k}\mathbf{k}'}|^2 + 2J_z^2 |\mathcal{F}^{z,\nu\nu'}_{\mathbf{k}\mathbf{k}'}|^2\Big), \tag{11}$$

which is the result presented in Eq. 2 and 3 of the main text.

II. ON THE SHAPE FACTORS $\mathcal{S}_i$

Here, first, for completeness, we recall the essential results from the Bloch-Wangness-Redfield (BWR) theory; and second, we further motivate the approximation proposed for the shape factors $\mathcal{S}_i$. The BWR theory analyzes the relaxation phenomena in an ensemble of magnetic moments subject to a random magnetic field $H_j(t)$, with $j = x, y, z$[3]. Considering the external static field H along $\hat{z}$ and the external radiofrequency field H_{rf} within the xy plane, the relaxation time results

$$\frac{1}{\tau_z} = \gamma^2 \Big(\tau_0 \overline{H}_z^2 + \frac{1}{2}\frac{\tau_0}{1+\omega_L^2\tau_0^2}(\overline{H}_x^2 + \overline{H}_y^2)\Big). \tag{12}$$

Here, τ_0 and $\overline{H}_j^2$ characterize the correlation in time of the random forces, $\overline{H_j(t)H_j(t+\tau)} = \overline{H_j}^2 \exp(-\tau/\tau_0)$. The asymmetric appearance of the different field components in Eq. 12 can be traced to their different role, which roughly speaking can be thought of as randomizing the Larmor frequency (the z-component) or the radiofrequency field (the xy components).

In our case, the random magnetic fields are given by the exchange with conduction electrons [Eqs. 3,4] and τ_0 can be related with the frequency of the spin fluctuations of these electrons, $\tau_0 \sim 1/\omega_{fl}$. As shown in Section **(I)**, while these random magnetic fields naturally depend on the crystal momentum of the carriers, their net effect on the relaxation rate is encoded in integral properties of the band structure –the shape factors $\mathcal{S}_i$. A comparison of Eqs. 2 and 3 of the main text with Eq. 12 suggests the identification $\tau_0\overline{H}_j^2 = TJ_j^2\mathcal{S}_j^2$, which provides a simple physical interpretation: the effective magnetic field exerted by the conduction electronic cloud over the timescale τ_0 is determined by the s-d exchange, the temperature and the shape factors $\mathcal{S}_i$.

Our DFT results indicate that due to the spin-orbit coupling the electronic structure, e.g. the density of states, presents a strong sensitivity to the direction of the Mn magnetic moments $\hat{m}$. The ansatz for the shape factors proposed in the main text aims to describe how such sensitivity can in turn affect the local moment relaxation process. Notice that strictly speaking, such effects are absent in Eq. 11 since correlations between the conduction electrons and the magnetic moments were neglected.

While a complete theory is beyond the scope of this work, a heuristic motivation for the ansatz presented in the main text is based on the following considerations. First, one considers that due to the spin-orbit coupling the instantaneous magnetic field exerted by the conduction electron cloud on the Mn moments depends on $\hat{m}$. Defining an instantaneous shape factor $s_j(t, \hat{m})$, one has

$$TJ_j^2\mathcal{S}_j^2 = TJ_j^2 \int_0^{\tau_{ce}} dt\, s_j(t, \hat{m}), \tag{13}$$

where τ_{ce} is the timescale associated with conducting electrons, typically $\sim \hbar/W$, with W their bandwidth. Second, the timescale of Mn spin fluctuations is assumed to be *larger* than τ_{ce} and naturally smaller than τ_z. In this intermediate timescale local rotations of $\hat{m}$ are followed essentially instantaneously by the electron cloud. Third, out of the ensemble of fluctuating Mn spins, a given configuration of H and $H_{\rm rf}$ picks up only certain components of the dynamical magnetization. For instance, for $H||c$ and $H_{\rm rf}||ab$, the measured relaxation originates in the in-plane component of the dynamical magnetization. Thus, we assume the electron cloud near the contributing Mn moments – and, in turn, the exerted magnetic field – to be better described by the electronic structure with in-plane magnetic moments.

Last, an argument for the estimation of the shape factor as the DOS computed for different $\hat{m}$ is based on considering the effects of the SOC perturbatively. At zero order, $\mathcal{S}_i^2 \propto D^2(\varepsilon_F)$. At first order, the matrix elements in $|\mathcal{F}_{\mathbf{kk}'}^{i,\nu\nu'}|^2$ in Eq. 11 are unaffected and the only effect on $\mathcal{S}_i^2$ is caused by the change in the eigenenergies. This change reflects in the electronic DOS in a manner that depends on the magnetic moment orientation. Our ansatz, $S_i^2 \propto D^2(\varepsilon_F)|_{\hat{m}\perp H}$, can be thought of as using this first-order perturbation theory result replacing the DOS with the one computed with DFT for different $\hat{m}$.

III. DETAILS OF THE DFT RESULTS

A. On the different electronic structure anisotropy of $MnBi_2Te_4$ and $MnBi_4Te_7$

To further understand the origin of the different sensitivity of the density of states of $MnBi_2Te_4$ and $MnBi_4Te_7$ to the orientation of the magnetic moments, here we consider the projection of the density of states on different atoms in the unit cell. For brevity, we define $\Delta = D_{\mathbf{m}||ab} - D_{\mathbf{m}||c}$. The key structural difference between these compounds is the additional quintuple layer (QL) in $MnBi_4Te_7$. One could, therefore, suspect that the contribution to the Bloch states of the atoms in the QL could be less sensitive to $\hat{m}$ than the contribution of the atoms in the septuple layer (SL). The additional QL would, therefore, act to reduce (by diluting) the overal sensitivity of the total density of states. Fig. 2(a) shows the projection of Δ on the QL (named $\Delta^{\rm QL}$) and on the SL ($\Delta^{\rm SL}$) and it can be seen that in the range of energies $[0.1, 0.3]$ eV, $|\Delta^{\rm QL}| < |\Delta^{\rm SL}|$. The difference are, however, rather modest. More significant than this is the effect that the additional QL has on the sensitivity of the SL contribution. To illustrate this, Fig. 2(b) shows the projection of Δ on the Bi atoms in the SL, both for $MnBi_2Te_4$ ($\Delta^{\rm Bi}_{124}$) and $MnBi_4Te_7$ ($\Delta^{\rm Bi}_{147}$). It can be seen that $|\Delta^{\rm Bi}_{124}|$ is significantly larger than $|\Delta^{\rm Bi}_{147}|$. A similar reduction is found in the projection on the Te atoms in the SL (not shown).

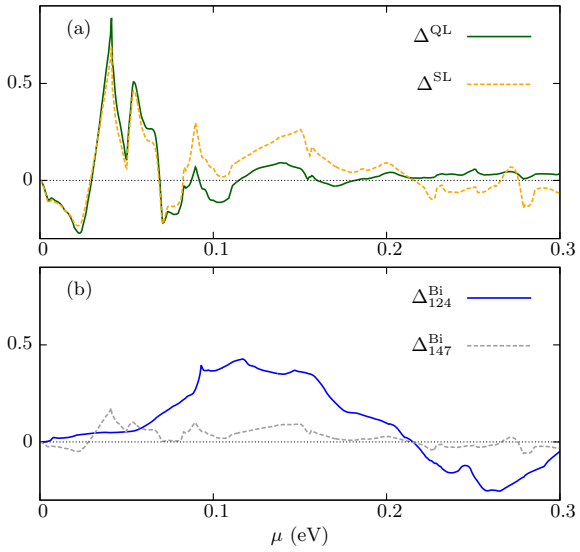

FIG. 1. Δ is the difference between the density of states corresponding to the cases $\hat{m}||ab$ and $\hat{m}||c$. (a) For $MnBi_4Te_7$, projection of Δ on the quintuple and sextpule layers. (b) For $MnBi_2Te_4$ and $MnBi_4Te_7$, projection of Δ on the Bi atoms in the septuple layer.

B. Carriers density vs Fermi energy.

For the estimation of τ_c/τ_{ab} as a function of the carrier density n, we consider the involved DOS at fixed n. For this, we first compute n as a function of the Fermi energy ε_F. These curves, shown in Fig. 2 also make very transparent the sensitivity of the electronic structure to the direction of the magnetic moments $\hat{m}$. While both compounds present a strong anisotropy at sufficient small doping, the main difference lies in the level of doping required to make the anisotropy negligble. While for $MnBi_4Te_7$ this occurs at small doping ($\mu \sim 50$meV, $n \sim 5 \times 10^{19}$ electron/cm^3), for $MnBi_2Te_4$ the anisotropy is preserved in a broader range of doping, which includes all estimates based on Hall-data reported in the literature.

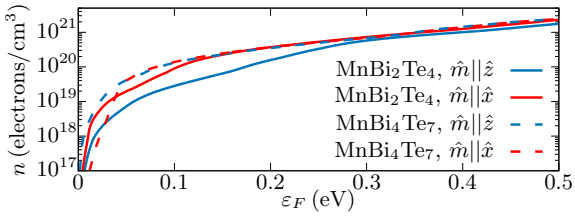

FIG. 2. Carrier density n vs Fermi energy ε_F for $MnBi_2Te_4$ and $MnBi_4Te_7$, for different orientations of the magnetic moments.

C. Density of states anisotropy in the FM phase

Fig. 3 shows the density of states of $MnBi_2Te_4$ and $MnBi_4Te_7$ in the ferromagnetic configuration. The same trends as in the antiferromagnetic configuration considered in the main text can be observed: larger density of states at small energies for in-plane configuration and overall smaller anisotropy in $MnBi_4Te_7$.

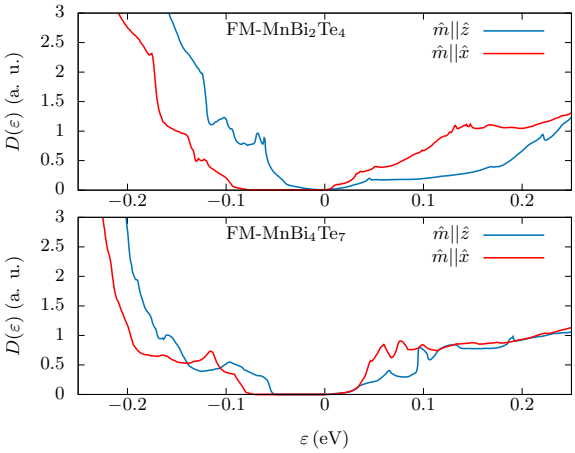

FIG. 3. Density of states vs energy for $MnBi_2Te_4$ and $MnBi_4Te_7$, for a ferromagnetic configuration. These calculations were performed with a k-mesh of $48 \times 48 \times 48$ ($MnBi_2Te_4$) and $48 \times 48 \times 8$ ($MnBi_4Te_7$) subdivisions in the Brilloin zone.

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[1] T. Moriya, "The effect of electron-electron interaction on the nuclear spin relaxation in metals," Journal of the Physical Society of Japan **18**, 516–520 (1963).

[2] R. E. Walstedt and A. Narath, "Relaxation of local-moment nuclei in metals," Phys. Rev. B **6**, 4118–4125 (1972).

[3] C.P. Slichter, *Principles of Magnetic Resonance*, Springer Series in Solid-State Sciences (Springer Berlin Heidelberg, 1996).