

Magnetic interactions and excitations in SrMnSb₂

Zhenhua Ning ¹, Bing Li ¹, Weilun Tang,¹ Arnab Banerjee ², Victor Fanelli ³, Douglas L. Abernathy ³,
Yong Liu,¹ Benjamin G Ueland ^{1,4}, Robert J. McQueeney,^{1,4} and Liqin Ke^{1,*}

¹Ames National Laboratory, U.S. Department of Energy, Ames, Iowa 50011, USA

²Department of Physics and Astronomy, Purdue University, West Lafayette, Indiana 47906, USA

³Neutron Scattering Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, USA

⁴Department of Physics and Astronomy, Iowa State University, Ames, Iowa 50011, USA



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The magnetic interactions in the antiferromagnetic (AFM) Dirac semimetal candidate SrMnSb₂ are investigated using *ab initio* linear response theory and inelastic neutron scattering (INS). Our calculations reveal that the first two nearest in-plane couplings (J_1 and J_2) are both AFM in nature, indicating a significant degree of spin frustration, which aligns with experimental observations. The orbital resolution of exchange interactions shows that J_1 and J_2 are dominated by direct and superexchange, respectively. In a broader context, a rigid-band model suggests that electron doping fills the minority spin channel and results in a decrease in the AFM coupling strength for both J_1 and J_2 . To better compare with INS measurements, we calculate the spin-wave spectra within a linear spin-wave theory, utilizing the computed exchange parameters. Although the calculated spin-wave spectra somewhat overestimate the magnon bandwidth, they exhibit overall good agreement with measurements from INS experiments.

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I. INTRODUCTION

Transition-metal dichalcogenides have the potential for intertwined charge, structural, and magnetic states [1]. In particular, the AMnX₂ (112), A = Ca, Sr, Ba, Eu, or Yb and X = Sb or Bi, have generated intense contemporary interest because they exhibit perfect or slightly distorted square magnetic Mn layers predicted to support Dirac or Weyl fermions [2–6]. The coupling of magnetic order or fluctuations to such topological quasiparticles is compelling as magnetic control may allow for tuning of topological properties. Thus, understanding the basic magnetic interactions in the 112 compounds is important.

The Dirac semimetal candidate SrMnSb₂ features a slightly distorted tetragonal structure and exhibits C-type antiferromagnetic (AFM) ordering, as shown in Figs. 1(a) and 1(b). The C-type order consists of Néel-type AFM order within the slightly distorted square Mn sublattices and ferromagnetic (FM) along the out-of-plane direction between them. The magnetic interactions and excitations of this compound have been studied using a combination of inelastic neutron scattering (INS) and density functional theory (DFT) [7]. The study considered the nearest-neighbor (NN) exchange coupling parameter J_1 , obtained from total energy mapping of various magnetic configurations. However, due to the semi-metallic nature of SrMnSb₂, it is reasonable to expect that the magnetic coupling extends to further neighbors.

To provide a more comprehensive picture, more recent work on various systems has expanded to the minimalistic Heisenberg J_1 - J_2 model [8,9], which factors in the

next-nearest-neighbor (NNN) coupling J_2 . This model offers an elegant and straightforward framework for interpreting various spin configurations resulting from the competition between J_1 and J_2 , taking into account their relative amplitudes and signs. Experimentally, valuable information about the values of SJ and the ratio of J_1/J_2 can be obtained through INS. However, further theoretical investigation is desirable, as it can offer a microscopic understanding of these interactions and guide the manipulation of J_{ij} , where J_{ij} is the parameter corresponding to the exchange coupling between sites i and j , to either alleviate spin frustration and tune magnetic order, or promote spin frustration and quantum spin fluctuations that can mediate electronic pairing.

Ab initio estimations of exchange parameters can be performed using different methods, including total energy mapping and linear response theory [10,11]. The total-energy-mapping method is widely employed due to its simplicity. For isotropic exchanges, the total energies of various collinear spin configurations are often calculated in first-principles methods and mapped onto a model spin Hamiltonian. Interactions beyond the isotropic exchanges, such as the off-diagonal part or the anisotropic diagonal part of the 3×3 exchange tensor J , can become important for more complicated materials systems that have large spin-orbit coupling (SOC) and/or broken inversion symmetry [12]. To estimate the relativistic-effect-originated exchanges, such as anisotropic exchange (anisotropic diagonal part of J tensor) or the Dzyaloshinskii-Moriya interaction (antisymmetric off-diagonal part of J tensor), SOC needs to be included in *ab initio* calculations, and noncollinear spin configurations need to be considered. However, the mapping method has limitations. For example, extracted interaction parameters can be nonunique due to the dependence on assumed model Hamiltonians. More-

*liqinke@ameslab.gov

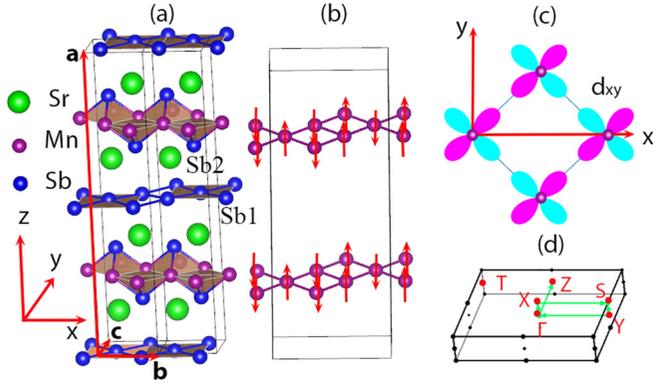


FIG. 1. Crystal structure and corresponding Brillouin zone of SrMnSb_2 . (a) Schematic representation of the crystal structure of SrMnSb_2 . The primitive unit cell is doubled along the \mathbf{b} direction to better illustrate the Mn grid. Sr, Mn, and Sb atoms are represented by green, purple, and blue spheres, respectively. The lattice vectors \mathbf{a} , \mathbf{b} , and \mathbf{c} are highlighted in red. For convenience in discussing the orbital contributions to exchange coupling, we align the longest lattice vector \mathbf{a} along the \hat{z} direction, and the nearly square Mn sublattice is on the \mathbf{bc} -basal plane, or equivalently, the xy plane. (b) C-type AFM ordering of spin moments of Mn atoms in the cell shown in (a). (c) Top view of d_{xy} orbitals for the Mn net shown in (b). (d) The first Brillouin zone of SrMnSb_2 with high-symmetry k points marked by red dots and k paths marked by green arrows. $X = [\frac{1}{2}, 0, 0]$, $Y = [0, \frac{1}{2}, 0]$, and $Z = [0, 0, \frac{1}{2}]$ in orthorhombic reciprocal lattice units (r.l.u.).

over, its applicability is limited in itinerant systems when the variability of on-site spin moments across different configurations, particularly in metallic compounds, becomes significant.

On the other hand, linear response theory evaluates the energy variations resulting from infinitesimal spin rotations away from the ground state. It is more computationally demanding but, in principle, more suitable for the calculation of low-temperature spin-wave (SW) excitations, which can be regarded as a small perturbation to the ground state. The linear response method is also easily extensible to beyond-DFT methods that may be challenging to access the total energy, such as many-body-perturbation-theory-based *GW* methods [13], to better describe electronic structures and magnetic interactions in various materials [11,14,15]. Finally, the linear response method allows for easy resolution of pairwise interactions into orbitals and band-filling effects, revealing the microscopic origin of these exchange couplings and providing guidance for band-structure engineering using doping and pressure.

Here, we apply linear response theory to investigate the magnetic interactions and excitations in SrMnSb_2 . We calculate and resolve isotropic exchange couplings, gaining insight into the microscopic origin of the magnetism. Using the obtained exchange parameters, we calculate the SW spectra via linear spin-wave theory (LSWT) and compare them with INS measurements. This comparison confirms the substantial magnetic frustration existing in the Mn layers. The linear-response results allow a discussion of how to tune the magnetic frustration by carrier doping.

II. COMPUTATIONAL AND EXPERIMENTAL METHODS

We first construct the real-space scalar-relativistic tight-binding (TB) Hamiltonian $H(\mathbf{R})$ using the maximally localized Wannier functions (MLWFs) method [16–18]. The reciprocal-space Hamiltonian $H(\mathbf{k})$ is obtained through Fourier transform. To better address the potential influences from the slightly distorted tetragonal structure, we symmetrize the TB Hamiltonian to ensure that the orthorhombic crystal symmetry is rigorously satisfied. Afterward, the corresponding Green's function $G(\mathbf{k}, \omega)$ is constructed for use in the linear response approach, as implemented in our recently developed TB Green's function code [19], to calculate the exchange couplings. Finally, with the exchange parameters in hand, we proceed to construct SW spectra using a LSWT.

A. Crystal structure

SrMnSb_2 crystallizes in the orthorhombic SrZnSb_2 -type ($Pnma$, space group no.62) structure with lattice parameters $a = 23.19 \text{ \AA}$, $b = 4.42 \text{ \AA}$, and $c = 4.46 \text{ \AA}$ [20]. The primitive cell contains four formula units (f.u.) and all atoms occupy different sets of $4c$ (m) sites. The crystal structure is shown in Fig. 1(a). For convenience in discussing the Mn-3d orbital contributions to the magnetic properties, we align the longest lattice vector \mathbf{a} along the \hat{z} direction, so the magnetic Mn layers are in the xy (\mathbf{bc}) basal plane. We also double the unit cell along the \mathbf{b} direction to better illustrate the Mn grid. The slight distortion of the \mathbf{bc} -basal plane results in the NNN exchange coupling J_2 becoming anisotropic with respect to the \hat{x} (\mathbf{b}) and \hat{y} (\mathbf{c}) directions, denoted as J_{2b} and J_{2c} , respectively.

There are two Sb sublattices, denoted in Fig. 1(a) as Sb_1 and Sb_2 , respectively. Two Sb_2 layers sandwich a Mn layer, forming MnSb_4 tetrahedra. The Sr atoms are relatively weakly bonded to the Sb_1 atoms and staggered above and below the Sb_1 layer, forming rhombus nets [21]. The C-type magnetic ordering lowers the crystal symmetry and separates the otherwise four equivalent Mn atoms into two sublattices, each possessing the same on-site magnetic moment but opposite orientation, as shown in Fig. 1(b).

Figure 1(c) illustrates Mn- d_{xy} orbitals pointing along the nearest neighbor direction, while Fig. 1(d) shows the first Brillouin zone (BZ) with special k points denoted. Due to the large separation between Mn layers, the super-superexchange between Mn layers is much weaker than the intralayer coupling, similar to the magnetic topological insulator MnBi_2Te_4 [22]. Such quasi-2D magnetic structures generally require beyond-mean-field approaches to estimate critical temperatures [23]. Experimental lattice constants and atomic position parameters [20] are used for all calculations in this work.

B. DFT details and TB Hamiltonian symmetrization

DFT calculations are performed using the generalized-gradient approximation (GGA) with the Perdew-Burke-Ernzerhof exchange-correlation parametrization [24] and the projector augmented wave approach [25,26] as implemented in the Vienna *ab initio* simulation package (VASP). To facilitate our subsequent analyses, we construct the MLWFs through a postprocessing procedure [16–18], as implemented in WANNI90 [27], using the output of the self-consistent

scalar-relativistic DFT calculation. In total, the TB basis comprises 44 MLWFs, encompassing $3d$ orbitals for four Mn atoms and $5p$ orbitals for eight Sb atoms in the unit cell. To minimize the spread functional for entangled energy bands, we adopt a two-step procedure [17]. For each spin channel, a real-space Hamiltonian $H^\sigma(\mathbf{R})$ with dimensions 44×44 is constructed to accurately represent the band structures in a specified “frozen” energy window near E_F . The energy bands are recalculated within TB to ensure that DFT bands can be accurately reproduced before further magnetic property calculations.

The process of wannierization for DFT bands may not always preserve the symmetry of wave functions and orbital characteristics. As we are also interested in understanding the potential impacts of the slightly distorted square Mn grid on exchange couplings, we symmetrize the Hamiltonian accordingly using

$$H_{ij,\sigma}^{\text{sym}}(\mathbf{R}) = \frac{1}{|\mathcal{G}_H|} \sum_{\mathcal{R} \in \mathcal{G}_H} \langle \mathbf{0} | \hat{\mathcal{R}} \hat{H} \hat{\mathcal{R}}^\dagger | \mathbf{R} \rangle_{ij,\sigma}, \quad (1)$$

where $\mathbf{0}$ denotes the central unit cell, \mathbf{R} denotes primitive translation vectors, i and j denote two sites within the primitive cell, σ is the spin channel, and \mathcal{R} represents symmetry operations within the subgroup \mathcal{G}_H of the Hamiltonian. A detailed implementation of Hamiltonian symmetrization can be found in Appendix A.

Before delving into magnetic properties calculations, we ensure alignment between the band structures generated by DFT and those produced by the symmetrized TB Hamiltonian:

$$H_{ij}^\sigma(\mathbf{k}) = \sum_{\mathbf{R}} H_{ij}^\sigma(\mathbf{R}) e^{-i\mathbf{k} \cdot \mathbf{R}}, \quad (2)$$

where \mathbf{k} denotes wave vectors.

C. Exchange couplings, spin wave, and critical temperature

The static linear response method with the long-wave approximation [10], based on Green’s function technique, has long been developed to calculate exchange couplings as defined in a Heisenberg-type Hamiltonian:

$$H = - \sum_{i \neq j} J_{ij}^e \hat{\mathbf{e}}_i \cdot \hat{\mathbf{e}}_j. \quad (3)$$

Here, J_{ij}^e is the isotropic exchange interaction parameter between sites i and j in the crystal, and $\hat{\mathbf{e}}_i$ is the unit vector pointing along the direction of the local spin moment at site i in the reference spin configuration. The early implementations utilized local-basis DFT methods, such as the linear muffin-tin orbital method [28], employing the atomic sphere approximation (ASA) and based on Green’s function technique. These methods provided highly efficient and useful descriptions of magnetic interactions in various systems, particularly those with close-packed structures. For example, they have been successfully applied to systems such as $\text{K}_2\text{Fe}_{4+x}\text{Se}_5$ [29], $T_2\text{AlB}_2$ ($T = \text{Fe}, \text{Mn}, \text{Cr}, \text{Co},$ and Ni) and their alloys [30], and $R(\text{Fe}_{1-x}\text{Co}_x)_{11}\text{TiZ}$ ($R = \text{Y}$ and Ce ; $Z = \text{H}, \text{C},$ and N) [31]. Additionally, more accurate full-potential methods have also been implemented to evaluate J_{ij} from the inverse static transverse susceptibilities, using the rigid-spin approximation

that projects susceptibility onto the spin density of the magnetic sites [11, 15, 32]. Lastly, modern plane-wave-based DFT methods have often been interfaced with the MLWFs method to generate realistic TB Hamiltonians that accurately describe the band structures within an energy window typically near the Fermi level (E_F) of a material. This approach also offers an attractive means to evaluate and analyze band structures and various other properties [33–35], with both efficiency and accuracy.

We carry out the linear response calculations using our recently developed TB code, which has been employed to efficiently analyze band structures [36, 37], Fermi surface [38], and magnetocrystalline anisotropy [19]. Starting from the TB Hamiltonian $H(\mathbf{k})$, we construct intersite Green’s function $G_{ij}^\sigma(\mathbf{k})$ on a $16 \times 16 \times 4$ k mesh to compute the exchange parameters $J_{ij}^e(\mathbf{q})$. Their orbital-resolved components $J_{ij,m}^e$ are defined by

$$J_{ij,m}^e(\mathbf{q}) = -\frac{1}{4\pi} \Im \int_{-\infty}^{E_F} d\omega \int_{\text{FBZ}} d\mathbf{k} [\Delta_i(\mathbf{k}) G_{ij}^\downarrow(\mathbf{k}, \omega) \times \Delta_j(\mathbf{k} + \mathbf{q}) G_{ji}^\uparrow(\mathbf{k} + \mathbf{q}, \omega)]_{mm}, \quad (4)$$

where the exchange-splitting matrix $\Delta_i(\mathbf{k}) = H_{ii}^\uparrow(\mathbf{k}) - H_{ii}^\downarrow(\mathbf{k})$. The real-space exchange constants $J_{ij}^e(\mathbf{R})$ are then obtained through a subsequent Fourier transform.

It is important to note that the neutron scattering community often adopts a different convention, wherein the Heisenberg Hamiltonian is frequently defined in terms of the spin vector \mathbf{S} as follows:

$$H = \sum_{i < j} J_{ij}^N \mathbf{S}_i \cdot \mathbf{S}_j. \quad (5)$$

For clarity and to facilitate comparison between calculations and experiments, by comparing Eqs. (3) and (5), we have

$$J_{ij}^N = -\frac{8}{\mathbf{m}_i \mathbf{m}_j} J_{ij}^e. \quad (6)$$

Here, $\mathbf{m}_i = 2\mathbf{S}_i$ is the magnetic moment (with sign) on site i . A positive (negative) J_{ij}^N indicates AFM (FM) coupling. In contrast to J_{ij}^N , J_{ij}^e is defined with respect to the given spin configuration, and a positive (negative) J_{ij}^e indicates that the given ordering of the corresponding pair is stable (frustrated). The critical temperatures, Curie temperature for FM materials or Néel temperature for AFM materials, can be estimated within the mean-field approximation as $T_C = \frac{2}{3} J_0^e$, where $J_0^e = \sum_i J_{0i}^e$. However, the mean-field approximation tends to overstate the critical temperature, especially in quasi-2D magnetic structures with very weak interlayer coupling. All J_{ij}^N discussed hereafter, unless specified otherwise, refer to J_{ij}^N .

With the *ab initio* exchange parameters obtained, the SW spectra can be calculated by solving the equation of motion method or through the bosonization of the spin Hamiltonian with Holstein-Primakoff [39] or other transformations that typically retain the two-boson terms in LSWT [40]. In the latter approach, higher-order four-boson terms can be included [23] to account for magnon-magnon interactions at finite temperatures.

D. INS experiment and analysis

The INS experiment was conducted on the wide angular-range chopper spectrometer (ARCS) [41] at the Spallation Neutron Source at Oak Ridge National Laboratory. For this experiment, 19 pieces of single crystals with a total mass of 386 mg were coaligned. Using an optical microscope with polarized light, twin domains due to the slightly distorted orthorhombic unit cell, each rotated by 90° with respect to the other, were observed on single-crystal pieces. Therefore single crystals were treated as pseudotetragonal while using our x-ray Laue camera to achieve coalignment. The horizontal scattering plane was defined as (H, K, L) , where the momentum transfer $\mathbf{q} = H\mathbf{a}^* + K\mathbf{b}^* + L\mathbf{c}^*$ is in orthorhombic reciprocal lattice units (r.l.u.). The rocking scans of the coaligned assembly yielded full widths at half maximum of 3° . The measurements were performed with the incident neutron energy of $E_i = 50$ or 125 meV at a temperature of $T = 20$ K.

To analyze the INS data, we performed least-squares fits and simulations utilizing the software package PYLISW [42].

III. RESULTS AND DISCUSSIONS

We begin by calculating the total energies of SrMnSb_2 with various magnetic orderings, including FM as well as C-, G-, A-, and Stripe-type AFM configurations. Our calculations confirm that among all the configurations, the C-type AFM ordering exhibits the lowest energy.

The MLWFs included in the TB Hamiltonian are determined by investigating the band-structure characters near the E_F in DFT. According to DFT calculations, the Mn-3d states dominate in the vicinity of E_F and in the energy range of -4.0 - 2.5 eV in the majority spin channel and 0 - 1.5 eV in the minority spin channel, exhibiting a spin splitting of about 3.8 eV. The more dispersive Sb-5p states also contribute to the band structure in the relevant energy window. On the other hand, the unoccupied Sr-4d states are located approximately 3 eV above E_F and can be neglected. Consequently, the TB basis consisting of Mn-3d and Sb-5p Wannier orbitals provides a reasonable description of the electronic structure in the vicinity of E_F .

As noted above, the wannierization procedure, in general, may not preserve the system's symmetry. The basis Wannier functions might center at positions that deviate from the atomic centers, and their orbital characteristics may not be preserved. We observe that the nonsymmetrized TB Hamiltonian, based on MLWFs, slightly breaks symmetry of SrMnSb_2 . Therefore, we symmetrize the TB Hamiltonian using Eq. (1) before conducting magnetic property calculations.

A. Electronic band structure and magnetization for SrMnSb_2

Figure 2 compares the electronic band structures along the k -path Γ -Y-S-Z- Γ -X, calculated using the symmetrized TB Hamiltonian (in blue) and DFT (in red). The excellent agreement in this energy window allows for a reasonable description of magnetic properties using the TB Hamiltonian.

With five electrons in the 3d shell, Mn atoms have a nearly fully occupied majority spin channel and a slightly occupied minority spin channel, resulting in a large mag-

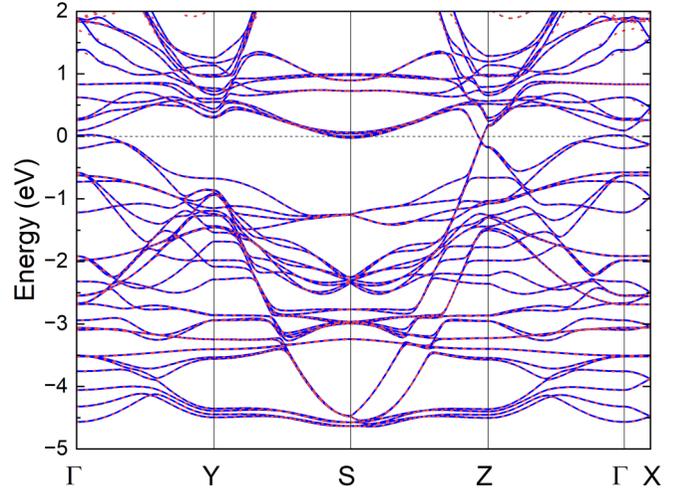


FIG. 2. Band structure of SrMnSb_2 calculated using tight-binding (solid blue lines) and density functional theory (red dots). They agree perfectly below 1 eV within this energy window. The horizontal dashed line denotes the Fermi level.

netic moment. The calculated Mn on-site magnetic moment of $3.81 \mu_B/\text{Mn}$ in DFT agrees well with the previous experimental value of $3.78 \mu_B/\text{Mn}$ [7,43], and the DFT values of $3.75 \mu_B/\text{Mn}$ to $3.78 \mu_B/\text{Mn}$ [7,44]. Furthermore, the symmetrized TB Hamiltonian gives $3.76 \mu_B/\text{Mn}$, agreeing well with the DFT and experimental results.

B. Magnetic interactions: Orbital contributions and band-filling effects

Figure 3 presents the real-space magnetic exchange parameters J_{ij} as a function of Mn-Mn distance R_{ij} obtained from the linear-response method. It is dominated by NN ex-

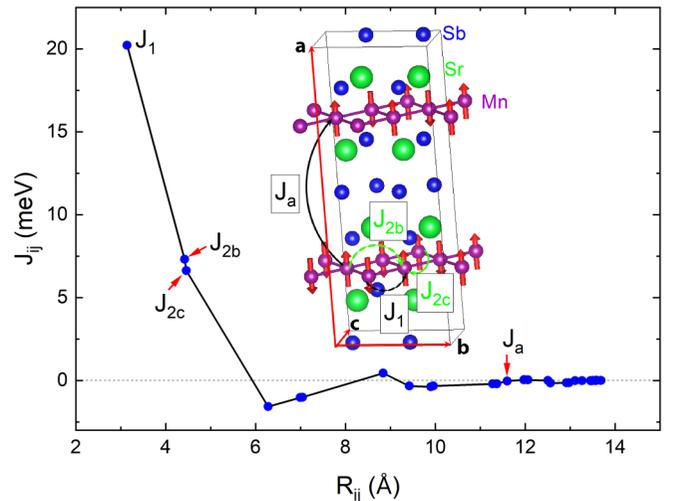


FIG. 3. Real-space magnetic exchange parameters J_{ij} , defined as J_{ij}^N in Eqs. (5) and (6), in SrMnSb_2 as a function of neighbor distance R_{ij} . The spin configuration of the C-type magnetic ground state, NN exchange coupling J_1 , NNN couplings J_{2b} and J_{2c} , and interlayer coupling J_a are denoted in the inset.

TABLE I. Pairwise exchange parameters J_{ij} (meV) in SrMnSb₂ for the Heisenberg Hamiltonian $H = \sum_{i<j} J_{ij} \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j$ as defined in Eq. (5). The NN and NNN exchange parameters J_1 , J_{2b} , and J_{2c} , and their corresponding distance R_{ij} and Mn-3d orbital resolutions are listed. As shown in Fig. 1(a), we align the Mn **bc**-basal plane in the xy Cartesian plane for the convenience of orbital-contribution discussion. Calculated SJ values are compared with corresponding INS values.

J_{ij}	No.	R_{ij}	xy	yz	z^2	xz	$x^2 - y^2$	Total	SJ	$SJ(\text{INS})$
J_1	4	3.14	8.36	3.68	3.18	3.85	1.60	20.67	38.86	27.51(6)
J_{2b}	2	4.42	0.77	1.47	0.84	4.19	0.21	7.48	14.06	11.19(5)
J_{2c}	2	4.46	0.83	3.51	0.77	1.49	0.17	6.77	12.73	7.78(9)

change coupling J_1 and NNN exchange coupling J_2 , showing a rapid decay as R_{ij} increases and becoming negligible after $R_{ij} = 8 \text{ \AA}$. The asymptotic behavior of fast decay suggests SrMnSb₂ a localized-moment system [32]. Both J_1 and J_2 exhibit positive values, indicating AFM interactions for both NN and NNN. The amplitude of J_2 is about 1/3 of J_1 and splits into two inequivalent couplings, J_{2b} and J_{2c} , with $R_{ij} = 4.42$ and 4.46 \AA , respectively, due to the slight structural distortion. Considering that the C-type configuration displays Néel-type in-plane AFM ordering, with AFM NN and FM NNN ordering within the **bc** basal plane, the sizable AFM J_2 suggests magnetic frustration. Table I summarizes the values of J_1 , J_{2b} , and J_{2c} , along with their Mn-3d orbital contributions, and corresponding values extracted from INS. The dominant contribution to J_1 originates from the d_{xy} orbital, which directly connects two NN Mn sites, as shown in Fig. 1(c). Other d orbitals make smaller AFM contributions to J_1 . Additionally, the d_{yz} and d_{xz} orbitals exhibit slightly different contributions, reflecting the small distortion in the Mn basal plane.

J_{2b} and J_{2c} are oriented along the **b** (\hat{x}) and **c** (\hat{y}) directions, respectively. Consequently, their primary contributions arise from d_{xz} and d_{yz} orbitals, respectively. An intriguing observation is that the $d_{x^2-y^2}$ orbital, which aligns directly with the coupling directions of J_{2b} and J_{2c} , contributes the least. This suggests that the direct exchange is relatively small due to the greater distance associated with J_2 and stands in stark contrast to the large contribution of d_{xy} to J_1 . Taken together, it becomes evident that the indirect superexchange through the Sb layers adjacent to the Mn layer plays a significant role in determining the magnitude of J_2 . Conversely, J_{2b} exhibits a slightly larger value than J_{2c} , possibly attributable to the former's shorter bond length compared to the latter, resulting from the subtle structural distortion.

The interlayer exchange couplings are notably weaker owing to the substantial distance ($R_{ij} > 11 \text{ \AA}$) between Mn layers along the **a** (\hat{z}) direction. The calculated nearest interlayer coupling J_a is FM, consistent with the experimental C-type spin ordering, and has a value of -0.029 meV , which is comparable with INS measurements as we discuss later.

As shown in Table I, in comparison to INS measurements, calculations somewhat overestimate the SJ values. Nonetheless, it is essential to highlight that the calculated trend in SJ values aligns with the experimental trend. This underscores the consistency between the theoretical predictions and experimental observations. The exchange interactions are also calculated by mapping multiple magnetic configurations to a minimalistic J_1 - J_2 - J_a Heisenberg model (see details in Ap-

pendix B). We found $J_1^{\text{eff}} = 22.47 \text{ meV}$, $J_2^{\text{eff}} = 5.25 \text{ meV}$, and $J_a^{\text{eff}} = -0.61 \text{ meV}$. Both J_1^{eff} and J_2^{eff} show good agreement with INS results and our linear response calculations. The values of J_1^{eff} and J_a^{eff} are consistent with those reported in a prior DFT study [7], whereas J_2^{eff} is approximately one order of magnitude larger than both the INS results and our linear response calculations. Overall, as we demonstrate later, the values of J_1 , J_2 , and J_2/J_1 calculated using the linear response theory exhibit better agreement with experimental data when compared to the mapping method. To explore the effects of electron or hole doping on exchange couplings and magnetic ordering, we calculate J_{ij} as functions of band filling within a rigid-band approximation. Figure 4(a) illustrates the dependence of the exchange parameters J_1 and J_2 on the number of doping electrons. (The corresponding orbital resolution can be found in the Appendix C.) With hole doping, both J_1 and J_2 increase slightly before decreasing. The energy range spanning from -0.5 eV to E_F is primarily governed by Sb-5p states in the density of states (DOS), with Mn-3d states contributing minimally, as depicted in Fig. 4(b). Consequently, the values of J_1 and J_2 exhibit only marginal alterations in the range of 0.1–0.45 hole/f.u. doping. The ratio of J_1 and J_2 is shown in Fig. 4(c), where the red dashed lines indicate the points where the square lattice reaches maximum frustration, residing on the boundary between Néel- and stripe-type AFM configurations within the basal plane. The J_1/J_2 ratio does not change significantly with up to 0.6 hole/f.u. doping. Furthermore, the resulting J_0^e increases slightly with weak hole doping, indicating a small increase in T_N . These computational results align well with a previous experimental study on hole doping [45], which found that the magnetic structure remains unchanged, maintaining the same in-plane Néel-type magnetic ordering with pronounced frustration, albeit with a slight increase in T_N .

Electron doping, on the other hand, has a much stronger effect on J_1 and J_2 ; doping within 0.1 electron/f.u. rapidly reduces their AFM coupling strength, with J_2 even transitioning to a weakly FM interaction. The resulting increase in the J_1/J_2 ratio indicates weaker spin frustration.

To understand the simultaneous decrease of J_1 and J_{2b} , we further resolve these parameters into orbitals and band-filling contributions. Figure 5 shows the resulting resolved $J_1(\omega)$ and $J_{2b}(\omega)$ along with the partial DOS projected on Mn-3d orbitals. Here, the band-filling-resolved exchange couplings, $J_{ij}(\omega)$, are defined as $J_{ij} = \int_{-\infty}^{E_F} d\omega J_{ij}(\omega)$, analogous to the density of states $D(E)$ and the total number of electrons $N = \int_{-\infty}^{E_F} d\omega D(\omega)$.

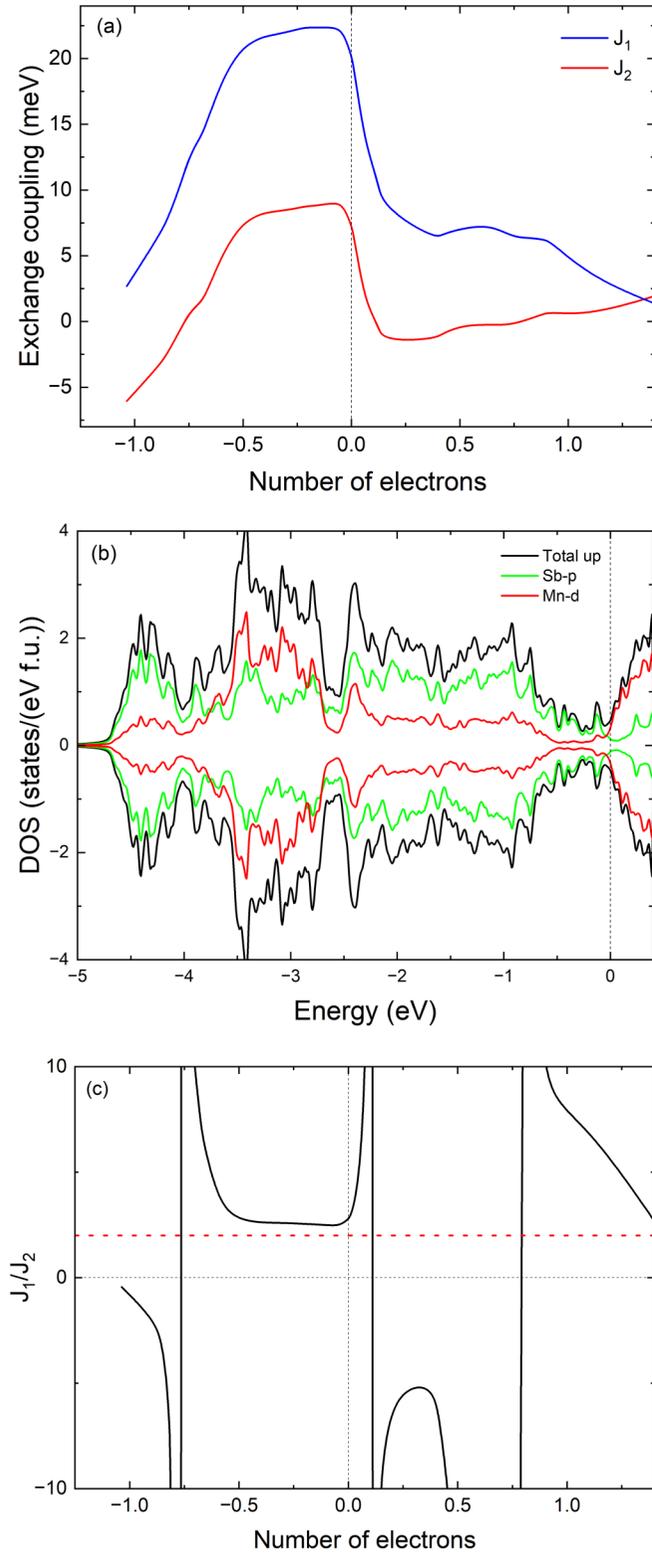


FIG. 4. Band-filling dependence of exchange parameters J_1 and J_2 , and density of states (DOS) in SrMnSb₂ calculated in TB. The Fermi level is shifted to zero. (a) J_1 and J_2 as functions of band-filling (Number of doping electrons per f.u.). (b) The total DOS, Sb-*p*, and Mn-*d* partial DOS (Number of states per f.u. per eV). (c) The ratio of J_1/J_2 as a function of doping, where the horizontal red dashed line at $J_1/J_2 = 2$ corresponds to maximum frustration.

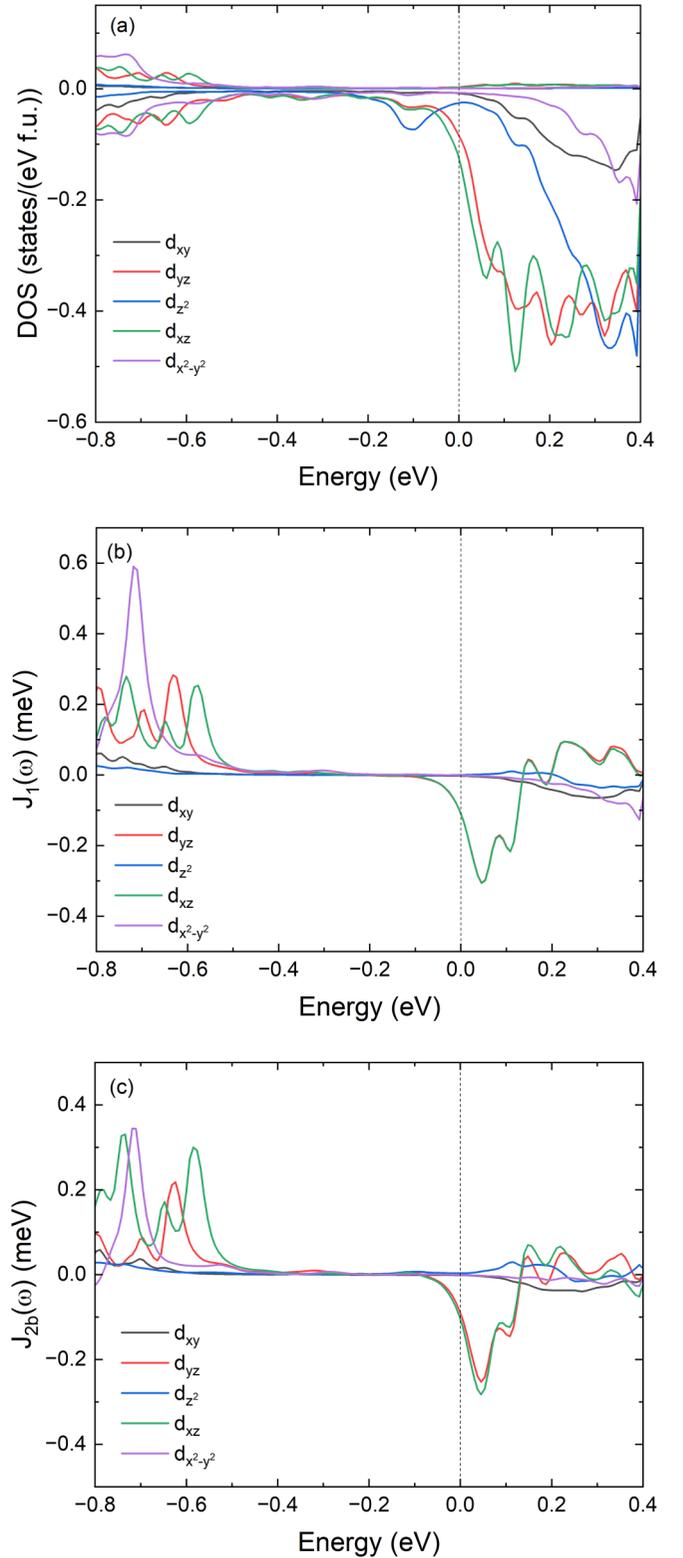


FIG. 5. (a) The partial DOS projected onto $3d$ orbitals of a single Mn site and orbital- and band-filling-resolved exchange couplings: (a) J_1 and (b) J_{2b} . The band-filling-resolved exchange couplings, such as $J_1(\omega)$, are defined as $J_1 = \int_{-\infty}^{E_F} d\omega J_1(\omega)$. $J_{2c}(\omega)$ behaves similarly to $J_{2b}(\omega)$ and is therefore not shown.

As depicted in Fig. 5(a), the energy window of -0.4 eV to 0.4 eV exhibits negligible DOS in the majority-spin channel, with E_F positioned at the shoulder of Mn- $d_{xz|yz}$ states in the minority spin channel. As shown in Figs. 5(b) and 5(c), in the energy range of -0.2 eV to 0.2 eV near E_F , all orbital-resolved $J_1(\omega)$ and $J_{2b}(\omega)$ values are mostly negative, indicating contributions to FM interactions. Conversely, for occupied states ranging from 0.6 eV to 0.8 eV below E_F , we have $J_1(\omega) > 0$ and $J_{2b}(\omega) > 0$, suggesting that the occupied states in this range contribute to AFM interactions for both J_1 and J_{2b} .

Therefore, in the rigid-band model, a slight hole doping (shifting E_F down to approximately 0.4 eV below E_F) would decrease the FM contribution, making J_1 and J_2 more AFM-like. In contrast, both J_1 and J_{2b} become less AFM (or more FM) with further hole doping, which removes the AFM contribution from states in the energy window of 0.5 – 0.8 eV below E_F , or with electron doping that adds FM contribution from states in the energy window of 0 – 0.2 eV above E_F . Notably, the band-filling dependence of both J_1 and J_{2b} primarily correlates with the filling of d_{xz} and d_{yz} states near E_F . Moreover, unsurprisingly, despite a substantial increase in the partial DOS above E_F for the out-of-plane Mn- d_{z^2} orbitals, its contribution to in-plane couplings is minimal.

C. Spin-wave spectra in J_1 - J_{2b} - J_{2c} - J_a model

Starting from the C-type ground state, we can derive the magnon dispersions of the J_1 - J_{2b} - J_{2c} - J_a model for SrMnSb₂ using the LSWT. The energies of the two magnon bands at $\mathbf{k} = H\mathbf{a}^* + K\mathbf{b}^* + L\mathbf{c}^*$ can be written as

$$\omega(\mathbf{k}) = M_0 \sqrt{(\alpha_k - d_0 \pm |\zeta_k|)^2 - |\beta_k|^2}, \quad (7)$$

where M_0 is the amplitude of the on-site Mn moment calculated in DFT, and

$$\begin{aligned} \alpha_k &= J_{2b} \cos(2\pi K) + J_{2c} \cos(2\pi L), \\ \beta_k &= \frac{1}{2} J_1 [1 + e^{i2\pi(K+L)} + e^{i2\pi K} + e^{i2\pi L}], \\ \zeta_k &= \frac{1}{2} J_a (1 + e^{i2\pi H}), \\ d_0 &= J_{2b} + J_{2c} + J_a - 2J_1. \end{aligned} \quad (8)$$

Figure 6 shows the magnon band structure along the Z - Γ - T path calculated using Eq. (7) with intralayer coupling parameters listed in Table I and interlayer coupling J_a . It is worth noting that Γ - Z [$(0, 0, \xi)$] corresponds to the J_{2c} direction, while Γ - T [$(0, -\xi, \xi)$] is slightly deviated from the J_1 direction. The inclusion of interlayer coupling J_a splits the magnon bands at Γ , as the acoustic and optical modes corresponding to the in- and out-of-phase spin precession with respect to the two Mn layers in the unit cell become energetically distinguishable. Although the calculation slightly overestimates the magnon bandwidth, the dispersions are in overall good agreement with INS measurements described below.

D. INS measurements and simulations

Figure 7 shows the INS data [(a)–(c)] and simulations using experimental parameters [(d)–(f)] of the SW spectra along three high-symmetry directions $(0, 1, L)(\Gamma - Z)$, $(0, 1/2 - K, 1/2 + K)(\Gamma - T)$, and

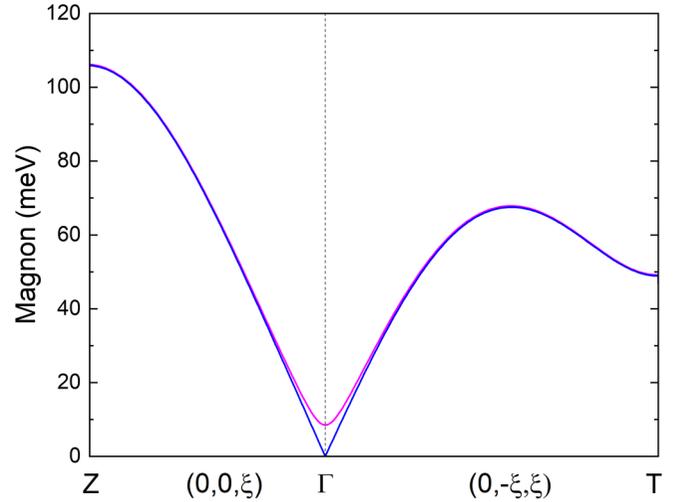


FIG. 6. Magnon band structure in SrMnSb₂ calculated using the J_1 - J_{2b} - J_{2c} - J_a model with parameters obtained from DFT. Magnetocrystalline anisotropy is not included. High-symmetry k points Y and T are illustrated in Fig. 1(d).

$(H, 1, 0)(\Gamma - X)$. The simulations are done through experimentally determined parameters as presented in Table I (or model 2 in Table II). Constant energy cuts of the data were taken and peaks in the cuts were fits to Gaussian functions in order to determine the peak center for a given E . The extracted peak centers of the energy line cuts are presented in Fig. 8. The data treatment and modeling and the cuts and fits are given in Sec. II of Ref. [46].

To understand the INS data, we calculate the SW spectra using LSWT with two models: one with (model 1) and the other without (model 2) enforcing $J_{2b} = J_{2c}$. The fitted single-

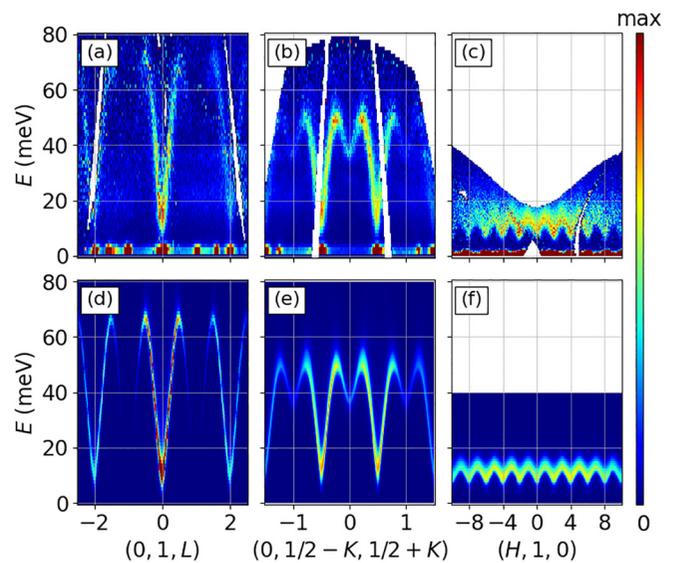


FIG. 7. INS data [(a)–(c)] and simulations using experimental parameters [(d)–(f)] of the SW spectra along three high-symmetry directions $[0, 1, L]$ ($\Gamma - Z$), $[0, 1/2 - K, 1/2 + K]$, and $[H, 1, 0]$ ($\Gamma - X$).

TABLE II. Parameters and single-ion anisotropy (SD) and magnetic interactions (meV) from two models.

Model	SD	SJ_1	SJ_{2b}	SJ_{2c}	SJ_a	$r\chi^2$
1	-0.26(6)	27.51(6)	9.36(2)	9.36(2)	-0.092(6)	15.04
2	-0.2559(9)	27.96(1)	11.19(5)	7.78(9)	-0.091(3)	4.79

ion anisotropy SD , in-plane magnetic interactions J_1 , J_{2b} , J_{2c} , and inter-layer interaction J_a are summarized in Table II.

In model 2, the difference between J_{2b} and J_{2c} induces asymmetry between the magnon bands along the path $(0, 1/2 - K, 1/2 + K)$, as shown in Fig. 8(b), resulting in higher energy for positive K values than negative K values. Additionally, the observed structural twins introduce a mixing of J_{2b} and J_{2c} . Due to the small difference between J_{2b} and J_{2c} and the relatively coarse energy resolution used for our measurements, the splitting is not observed in the INS data. Nonetheless, model 2 provides a slightly better fit to the dispersion with a smaller reduced χ^2 ($r\chi^2$) compared to model 1, as shown in Fig. 8 and Table II.

Overall, when compared to INS measurements, the SW dispersion derived from first-principles calculations, although somewhat overestimating the bandwidth, agrees reasonably well with INS measurements. Correspondingly, the intralayer interactions (J_1 , J_{2b} , and J_{2c}) as well as the interlayer interaction (J_a) are comparable to values extracted from INS. The concordance between the theoretical predictions and the experimentally derived SJ -based calculations affirms the accuracy of our first-principles methodology in effectively predicting magnetic interactions within the materials.

The overestimation of SJ values in our calculations could potentially be addressed. For instance, the MLWFs we used here typically have tails extending into other sites, which may blur the definition of calculated exchange couplings between two sites. It would be worthwhile to investigate whether a representation of the on-site moment in a basis that better localizes the magnetic moment within the atomic

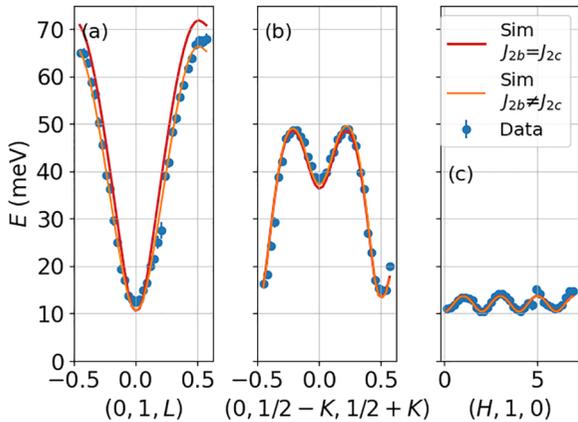


FIG. 8. Magnon dispersion along three high-symmetry directions determined from fits to the INS data. Lines show fits to the dispersion using the two different model Hamiltonians discussed in the text. Model 1 with $J_{2b} = J_{2c}$ is shown as red solid line while model 2 with $J_{2b} \neq J_{2c}$ is shown as orange solid line.

sphere could improve the description. Another consideration is the inclusion of electron-correlation effects beyond DFT. Typically, one may consider additional electron repulsion for the localized Mn-3d orbitals beyond DFT using methods such as DFT + U . This approach often promotes electron localization, thereby increasing the magnetic moment while decreasing the exchange coupling. Our preliminary results from applying an additional Hubbard-like U potential in DFT to Mn-3d orbitals show enhanced electron localization and reduced intersite exchange couplings, leading to a better agreement with experiments in terms of overall magnon bandwidth. However, ideally, the choice of the U values should be justified, for example, by comparing calculated and experimentally measured band structures. GW -based methods can be even more useful, as they are not only parameter-free but also contain nonlocal off-site exchange-correlations that do not exist in DFT + U but can be crucial for properly describing the superexchanges. A systematic study of electron correlation effects is beyond the scope of the present work.

IV. CONCLUSIONS

This study on SrMnSb₂ presents a comprehensive understanding of its electronic and magnetic properties, achieved through a synergy of theoretical methodologies and experimental data. The exploration of exchange coupling parameters in SrMnSb₂ is carried out using linear response theory within a realistic TB model. Our calculated intralayer interactions (J_1 , J_{2b} , and J_{2c}) and interlayer interaction (J_a) are in good agreement with INS results. Moreover, the study reveals a sizable AFM NNN exchange coupling J_2 , introducing significant spin frustration within the basal plane. Orbital-resolved contributions to exchange couplings help reveal the microscopic origin of the exchange interactions. Notably, we find that the NN exchange coupling J_1 in SrMnSb₂ predominantly arises from the contributions of Mn d_{xy} orbitals, aligning with the nearest Mn-Mn bond, while J_{2b} and J_{2c} receive primary contributions from d_{xz} and d_{yz} orbitals, respectively. This reflects the dominance of direct-exchange and superexchange nature for J_1 and J_2 , respectively. The band-filling dependence of exchange coupling based on the rigid-band model reveals that electron doping is expected to weaken both J_1 and J_2 while relieving spin frustration through increasing the J_1/J_2 ratio. Moreover, the magnetic structure of SrMnSb₂ is anticipated to remain unaltered under carrier doping, which is in agreement with previous hole-doping investigations [45]. Introducing additional electron correlation within the Mn-3d orbitals can promote electron localization and reduce the magnetic coupling, further improving the agreement with experiments. In summary, this investigation not only advances our comprehension of SrMnSb₂ but also underscores the efficacy of combining *ab initio* and experimental methods in elucidating intricate materials and their magnetic interactions.

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APPENDIX A: HAMILTONIAN SYMMETRIZATION

Wannier functions (WFs) are localized atomic-like orbitals defined at each atom site. For a set of isolated bands that are separated from all other lower and higher bands throughout the BZ by band gaps, its electronic states can be well described by a set of WFs $|w_{\mathbf{R}}^{\mu}\rangle$, where \mathbf{R} is the location of the unit cell considered, and μ is the index of WFs in the cell. For a multiple-site system, μ is a combination of quantum numbers n_i, l_i, m_i, σ_i , and site position vectors τ_i . For systems with only one site in the primitive cell, we have $\tau_i = (0, 0, 0)$.

When a WF is rotated by one point symmetry operation $\hat{\mathcal{R}}$ of the system, it can only transform into WFs belong to equivalent sites with the same quantum numbers n_i and l_i , that is, the symmetry can only mix states with different magnetic quantum numbers and spins

$$\hat{\mathcal{R}}|w_{\mathbf{R}+\tau_i}^{\beta_i m_i \sigma_i}\rangle = \sum_{m'_i \sigma'_i} \mathcal{R}_{m'_i \sigma'_i m_i \sigma_i}^{l_i} |w_{\mathbf{R}+\tau'_i}^{\beta_i m'_i \sigma'_i}\rangle, \quad (\text{A1})$$

where $\hat{\mathcal{R}}(\mathbf{R} + \tau_i) = \mathbf{R}' + \tau'_i$ and β_i the combination of n_i and l_i . According to the orthogonalization of WFs, the transformation matrix $\mathcal{R}_{\mathbf{R}\mathbf{R}'}$ with elements given by

$$\langle w_{\mathbf{R}+\tau'_i}^{\beta_i m'_i \sigma'_i} | \hat{\mathcal{R}} | w_{\mathbf{R}+\tau_i}^{\beta_i m_i \sigma_i} \rangle = \mathcal{R}_{m'_i \sigma'_i m_i \sigma_i}^{l_i} \delta_{\mathbf{R}+\tau'_i, \hat{\mathcal{R}}(\mathbf{R}+\tau_i)} \delta_{\beta_i, \beta'_i}. \quad (\text{A2})$$

It is obvious that the transformation matrix \mathcal{R} should be l and n diagonal.

The transformation matrix \mathcal{R} can be obtained by the behavior of WANNIER90's basis orbitals under symmetry operations. WANNIER90 uses real spherical harmonics $\mathcal{Y}_{l,m}$ which is related to complex spherical harmonics Y_l^m by

$$\mathcal{Y}_{l,m} = \begin{cases} \sqrt{2}(-1)^m \Re\{Y_l^m(\theta, \phi)\} & m > 0 \\ \sqrt{2}(-1)^m \Im\{Y_l^{-|m|}(\theta, \phi)\} & m < 0, \\ \Re\{Y_l^m(\theta, \phi)\} & m = 0 \end{cases}$$

$$= \begin{cases} \frac{1}{\sqrt{2}}[Y_l^{-|m|} + (-1)^m Y_l^{|m|}] & m > 0 \\ \frac{i}{\sqrt{2}}[Y_l^{-|m|} - (-1)^m Y_l^{|m|}] & m < 0. \\ Y_l^0 & m = 0 \end{cases} \quad (\text{A3})$$

This equation defines the unitary transformation matrix $\mathbf{U}_{\mathbb{C} \rightarrow \mathbb{R}}$ between $\mathcal{Y}_{l,m}$ and Y_l^m . The angular momentum operator \hat{L} in the basis of real spherical harmonics can then be written as

$$\hat{L}^{\mathbb{R}} = \mathbf{U}_{\mathbb{C} \rightarrow \mathbb{R}} \hat{L}^{\mathbb{C}} \mathbf{U}_{\mathbb{C} \rightarrow \mathbb{R}}^{\dagger} = \mathbf{U}_{\mathbb{C} \rightarrow \mathbb{R}} \hat{L}^{\mathbb{C}} \mathbf{U}_{\mathbb{R} \rightarrow \mathbb{C}}. \quad (\text{A4})$$

In the presentation of complex spherical harmonics Y_l^m , the nonvanished matrix elements of $\hat{L}^{\mathbb{C}}$ are

$$\begin{aligned} \langle l, m | \hat{L}_z | l, m \rangle &= \hbar m, \\ \langle l, m-1 | \hat{L}_- | l, m \rangle &= \hbar \sqrt{l(l+1) - m(m-1)}, \\ \langle l, m+1 | \hat{L}_+ | l, m \rangle &= \hbar \sqrt{l(l+1) - m(m+1)}. \end{aligned} \quad (\text{A5})$$

The rotation on orbital function is defined by

$$\hat{\mathcal{R}}^{\mathbb{C}} = e^{-i\hat{\mathbf{n}} \cdot \hat{L}^{\mathbb{C}} \phi}, \quad (\text{A6})$$

where $\hat{\mathbf{n}}$ is the axis of the rotation symmetry and ϕ the rotation angle. Then the transformation matrix \mathcal{R} for the orbital part can be obtained by

$$\hat{\mathcal{R}}^{\mathbb{R}} = \mathbf{U}_{\mathbb{C} \rightarrow \mathbb{R}} \hat{\mathcal{R}}^{\mathbb{C}} \mathbf{U}_{\mathbb{R} \rightarrow \mathbb{C}} = \mathbf{U}_{\mathbb{C} \rightarrow \mathbb{R}} e^{-i\hat{\mathbf{n}} \cdot \hat{L}^{\mathbb{C}} \phi} \mathbf{U}_{\mathbb{R} \rightarrow \mathbb{C}}. \quad (\text{A7})$$

The rotation on spinor can be written as

$$\hat{\mathcal{R}}^{\hat{\sigma}} = e^{-i\hat{\mathbf{n}} \cdot \hat{S} \phi}. \quad (\text{A8})$$

The spin angular momentum operator \hat{S} is defined as

$$\hat{S} = \frac{\hbar}{2}(\sigma_x, \sigma_y, \sigma_z), \quad (\text{A9})$$

where σ_x, σ_y , and σ_z are Pauli matrices given by

$$\sigma_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \quad \sigma_y = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}, \quad \sigma_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}. \quad (\text{A10})$$

Thus, in the representation of real spherical harmonics and spinor, the rotation is given by $\hat{\mathcal{R}} = \hat{\mathcal{R}}^{\mathbb{R}} \hat{\mathcal{R}}^{\hat{\sigma}}$.

The Hamiltonian \hat{H} of a system is invariant under its all symmetry operations

$$\hat{H} = \hat{\mathcal{R}} \hat{H} \hat{\mathcal{R}}^{\dagger}. \quad (\text{A11})$$

The elements of real-space Hamiltonian $H^{\mathbf{R}}$ are defined by $H_{ij}^{\mathbf{R}} = \langle w_{\tau_i}^{\beta_i m_i \sigma_i} | \hat{H} | w_{\mathbf{R}+\tau_j}^{\beta_j m_j \sigma_j} \rangle$ that under symmetry operations transforms as

$$\begin{aligned} H_{ij}^{\mathbf{R}} &= \langle w_{\tau_i}^{\beta_i m_i \sigma_i} | \hat{H} | w_{\mathbf{R}+\tau_j}^{\beta_j m_j \sigma_j} \rangle \\ &= \langle w_{\tau_i}^{\beta_i m_i \sigma_i} | \hat{\mathcal{R}} \hat{H} \hat{\mathcal{R}}^{\dagger} | w_{\mathbf{R}+\tau_j}^{\beta_j m_j \sigma_j} \rangle \\ &= \sum_{\mu'_i, \mu'_j, \mathbf{R}''} \langle w_{\tau_i}^{\beta_i m_i \sigma_i} | \hat{\mathcal{R}} | w_{\mathbf{R}+\tau'_i}^{\beta'_i m'_i \sigma'_i} \rangle \langle w_{\mathbf{R}+\tau'_i}^{\beta'_i m'_i \sigma'_i} | \hat{H} | w_{\mathbf{R}''}^{\beta'' m'' \sigma''} \rangle \\ &\quad \times \langle w_{\mathbf{R}''}^{\beta'' m'' \sigma''} | \hat{\mathcal{R}}^{\dagger} | w_{\mathbf{R}+\tau_j}^{\beta_j m_j \sigma_j} \rangle. \end{aligned} \quad (\text{A12})$$

Write in matrix form

$$\langle \mathbf{0} | \hat{\mathcal{R}} \hat{H} \hat{\mathcal{R}}^{\dagger} | \mathbf{R} \rangle = \sum_{\mathbf{R}''} \mathcal{R}_{\mathbf{0}\mathbf{R}''} H^{\mathbf{R}''-\mathbf{R}} \mathcal{R}_{\mathbf{R}''\mathbf{R}}. \quad (\text{A13})$$

If a Hamiltonian does not satisfy the system's symmetry, its symmetrized Hamiltonian can be obtained as the average over all symmetry operations given by

$$H^{\text{sym}}(\mathbf{R}) = \frac{1}{|\mathcal{G}_H|} \sum_{\mathcal{R} \in \mathcal{G}_H} \langle \mathbf{0} | \hat{\mathcal{R}} \hat{H} \hat{\mathcal{R}}^{\dagger} | \mathbf{R} \rangle, \quad (\text{A14})$$

where $|\mathcal{G}_H|$ is the number of symmetry operators in the group \mathcal{G}_H .

APPENDIX B: CALCULATE J_{ij} BY MAPPING TOTAL ENERGY CALCULATIONS

For SrMnSb₂ in a J_1 - J_2 - J_d model, to extract the three exchange parameters by energy-mapping method, we can calculate the total energies of four AFM magnetic configurations: C-, A-, G-, and Stripe(S)-type. A supercell with basis vectors \mathbf{a} , $\mathbf{b} + \mathbf{c}$ and $\mathbf{b} - \mathbf{c}$ is constructed and used to simulate these four magnetic configurations. According to the Heisenberg model as stated in Eq. (5), we obtain

$$\begin{aligned} 4E_A/S^2 &= 2J_1 + 2J_2 - J_c/2, \\ 4E_G/S^2 &= -2J_1 + 2J_2 - J_c/2, \\ 4E_S/S^2 &= -2J_2 - J_c/2, \\ 4E_C/S^2 &= -2J_1 + 2J_2 + J_c/2, \end{aligned} \quad (\text{B1})$$

where the energies E_A , E_G , E_{SA} , and E_C are the calculated total energy per magnetic atom for the four configurations. Here we do not distinguish J_{2b} and J_{2c} , instead, an average of them as J_2 is calculated. Then the exchange coupling constants are given by

$$\begin{aligned} J_c &= 4(E_C - E_G)/S^2, \\ J_1 &= (E_A - E_G)/S^2, \\ J_2 &= (E_A - E_S)/S^2 - J_1/2. \end{aligned} \quad (\text{B2})$$

APPENDIX C: ORBITAL RESOLVED EXCHANGE COUPLINGS

Orbital-resolved exchange couplings J_1 and J_2 of SrMnSb₂ as a function of Fermi energy are calculated through $J_{1,2} = \int_{-\infty}^{E_F} d\omega J_{1,2}(\omega)$, as illustrated in Fig. 9. The figure shows contributions from each d -orbital. The dominant contribution to J_1 originates from Mn- d_{xy} orbitals, while the dominant contribution to J_2 is from Mn- d_{xz} orbitals.

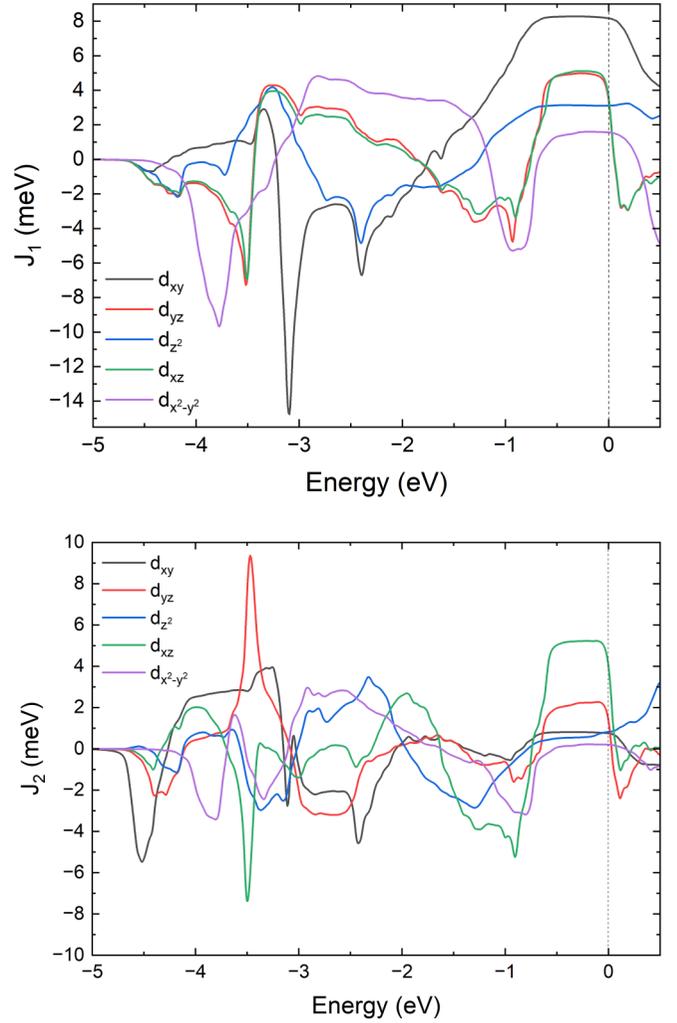


FIG. 9. Orbital-resolved exchange couplings of SrMnSb₂ as a function of Fermi energy are plotted with color weights, with black identifying the Mn- d_{xy} state, blue the Mn- d_{3z^2-1} states, red the Mn- d_{yz} states, green the Mn- d_{xz} state, and purple the Mn- $d_{x^2-y^2}$. The horizontal zero is the Fermi level.

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