Interplay of Magnetism and Topological Superconductivity in Bilayer Kagome Metals

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The binary intermetallic materials, M3Sn2 (M=3d transition metal) present a new class of strongly correlated systems that naturally allows for the interplay of magnetism and itinerant electrons. Using first principles calculations we confirm that bulk Fe3Sn2 is a ferromagnetic metal, and show that M=Ni and Cu are paramagnetic metals with non-trivial band structures. Focusing on Fe3Sn2 to understand the effect of enhanced correlations in an experimentally relevant setting of atomistically thin single bilayer, our ab-initio results show that dimensional confinement naturally exposes the flatness of band structure associated with the kagome geometry in a resultant ferromagnetic Chern metal. We use a multistage minimal modelling of the magnetic bands progressively closer to the Fermi energy to capture the physics of the Chern metal and find non-zero anomalous Hall response over a material relevant parameter regime along with a possible superconducting instability of the spin-polarised band resulting in a topological superconductor. Our calculations thereby indicate that Fe3Sn2 can host novel physics in the bilayer limit.

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Introduction—Accentuated quantum fluctuations due to dimensional confinement and electron-electron correlations are at the heart of some of the novel electronic phases of condensed matter—e.g., high temperature superconductivity in naturally layered cuprates[1] and iron pnictides[2]; the low dimensional frustrated magnets[3]; easily exfoliable materials such as graphene[4] and more recently twisted bilayer graphene [5]; and manufactured two dimensional (2D) electron gas leading to integer and fractional quantum Hall effects[6] as well as various synthetic heterostructures[7, 8].

An interesting recent addition to this ongoing research is the binary intermetallic series MmXn with M = 3d transition metals (TMs) forming stacked kagome layers, separated by X = Sn, Ge spacer layers in stoichiometric ratios of m:n=3:1, 3:2 or 1:1. A number of very interesting properties of these system have been reported recently that includes: Dirac-like electronic structure[9, 10], large anomalous Hall effect[11, 12] and magnetic Weyl excitations[13]. Diverging density of states, probed by scanning tunneling spectroscopy, have been reported for Fe3Sn2[14] as well as for the ternary kagome ferromagnetic compound, Co3Sn2S2[15], though the existence of flat band, a characteristic to Kagome geometry, has remained inconclusive possibly due to presence of other entangled bands in the 3D material. Very recently however, the bulk material FeSn has been synthesised[16] that contains stacked single-layer kagome planes and it has been claimed to have momentum space signatures of flat bands at energies few hundreds of meV below and above Fermi level. In this backdrop, it is curious to explore the consequence of dimensional confinement in these intermetallics by considering the atomistically thin limit, which is expected to further enhance the strong correlation physics. This would provide the exciting opportunity to possibly investigate the interplay of the flat-band physics and fluctuating magnetism of the low-dimensional 3d transition metal layers interacting with 2D spacer itinerant electrons.

In this paper, we explore the above possibility within the framework of first-principles density functional theory (DFT) and effective low energy minimal models inspired by the DFT band structure. To probe the effect of confinement, we consider one unit of kagome bilayer, sandwiched between two Sn layers (Fig. 5(b)), derived out of the M3Sn2 bulk structure (Fig. 5(a)). In addition to Fe compound which is already synthesized as a bulk material, we also consider two more late TM based compounds, namely Ni and Cu compounds, which are yet-to-be synthesized, to understand the generic behaviour of the above family of materials. The calculated cleavage energies[17, 18] show that the energy costs involved in creation of such bilayers are 1 to 2 J/m2, similar to that required for creating 2D MXenes, the 2D counterparts of MAX phases[19]. Given the successful synthesis of MXenes[20] through the chemical etching route, possible synthesis of pro-

FIG. 1: (a) Layered arrangement in bulk M3Sn2. (b) Bilayer M4Sn6 derived out of the bulk layered structure. (c) Stacking of two kagome layers within the bilayer, viewed along the out-of-plane direction.
posed bilayer structures is promising.

Analysis of the electronic structure of bulk $M_3Sn_2$ ($M=$Fe, Ni, and Cu) prompts us to conclude that Fe$_3$Sn$_2$ is the natural choice to search for fluctuation-driven physics in magnetic flat bands, as both Ni$_3$Sn$_2$ and Cu$_3$Sn$_2$ turn out to be nonmagnetic within our DFT calculations. Thus, while Ni$_3$Sn$_2$ and Cu$_3$Sn$_2$ have interesting band structure (see below) and hence deserve attention, Fe appears to be in a sweet spot of the interplay of correlations and band physics in the late 3d TM series. Our DFT calculations for bilayer Fe$_3$Sn$_2$ reveal that confinement to the bilayer limit, indeed causes drastic changes in the electronic structure, resulting in formation of near-flat bands within $\pm 10$ meV of Fermi level, together with massive Weyl-like band features. Interestingly, the ferromagnetic correlations in the 3D compound are found to survive down to bilayer limit, thus resulting in almost flat bands with non-zero Chern number and thereby realising a Chern metal in the bilayer system. Inclusion of magnetic fluctuation effects within the model calculations of the kagome bilayer show that fluctuation-driven topological superconductor is a possibility.

Electronic Structure of Bulk $M_3Sn_2$ — Rhombohedrally structured bulk $M_3Sn_2$ consist of bilayers of kagome network of metal atoms consisting of big and small equilateral triangles, sandwiched between stanene layers. The Sn' atoms approximately occupy the centre of the hexagons in the kagome plane while the kagome bilayers are separated by honeycomb Sn'' layers (Fig. 5(a)) [21]. The crystal structures of Ni$_3$Sn$_2$ and Cu$_3$Sn$_2$ are obtained by starting from the crystal structure of Fe$_3$Sn$_2$, replacing the Fe atoms by Ni and Cu respectively, and followed by fully relaxing the lattice constants and the atomic positions keeping the symmetry fixed. DFT calculations are performed in plane wave basis as implemented in Vienna Ab-initio Simulation Package (VASP),[22–24] with exchange-correlation functional within generalised gradient approximation (GGA)[25]. Also, correlation effect at TM sites is included within GGA+U[26] with $U = 0.5\text{ eV}$ [27]. Weak spin-orbit coupling (SOC) is also included for the TM 3d states which appears to be a crucial ingredient to drive the topological behaviour as well as stability of the magnetism in the bilayer limit (see below). Further details are provided in the Supplementary Material (SM)[28].

Figs. 2(a)–(c) shows the GGA+U+SOC band structure of Fe$_3$Sn$_2$, Ni$_3$Sn$_2$ and Cu$_3$Sn$_2$ plotted along the high symmetry directions of the hexagonal Brillouin zone (BZ).[29] Interestingly, while the DFT calculations for Fe compound stabilises a magnetic solution with moment $\approx 2.2\ \mu_B$ at Fe site, in agreement with reported experiments,[27] both Ni and Cu compounds, turned out to be nonmagnetic. Focusing on the corresponding density of states (DOS) (cf. Figs 2(d)–(f)), we find while the low-energy states are primarily dominated by peaked Fe d states with admixture from Sn p due the covalency for Fe$_3$Sn$_2$, the low-energy states for Ni$_3$Sn$_2$ show a much stronger admixture of Ni d and Sn p, and are dominated almost entirely by broad Sn p bands for Cu$_3$Sn$_2$. Employing the Stoner criteria of magnetism, as would be appropriate for metallic systems, we find $I \times N(E_F)$ ($I =$ Stoner parameter, $N(E_F) =$ DOS at $E_F$) to be larger than 1 (1.4) for Fe$_3$Sn$_2$, and significantly less than 1 for Ni$_3$Sn$_2$, and Cu$_3$Sn$_2$ (0.3 and 0.1, respectively), justifying presence of magnetism in the former compound and its absence in the latter compounds. The band structures of Fe$_3$Sn$_2$, which is in good agreement with published literature [27], and that of Ni$_3$Sn$_2$ and Cu$_3$Sn$_2$ show topologically non-trivial Dirac-points (encircled in Fig. 2(a–c)). The electronic structure of Ni$_3$Sn$_2$ and Cu$_3$Sn$_2$ compounds though appear interesting and deserves further attention, in keeping the focus on the interplay of magnetic fluctuations and topological properties in low dimensionality, we concentrate on the properties of Fe bilayer in the rest of this paper.

Electronic Structure of Bilayer FeSn — We focus on the Sn'' terminated Fe$_n$Sn$_6$ bilayers as shown in Fig. 5(b). The kagome bilayer (BL), which in this case is isolated, consists of two kagome layers of TM atoms, shifted with respect to each other. It is important to note that the bulk FeSn structure studied in recent literature[16] has single kagome layer separated from neighbouring kagome layers along c-axis by single Sn layer, and is different from present case.

The GGA+SOC+U band structure of Sn-terminated bilayer FeSn consisting of two kagome layers (cf. Fig. 5(c)) is shown in Fig. 3 along with the orbital characters of the bands projected to the Fe d character. DFT estimated intra- and inter-kagome layer magnetic interaction within the bilayer geometry turned out to be both ferromagnetic, as in bulk Fe$_3$Sn$_2$ compound, with estimated values of $\approx 10$ meV and 0.3 meV, respectively. This is in contrast to magnetic behaviour of bulk FeSn reported recently[16], where the ferromagnetic Fe kagome layers are coupled antiferromagnetically.

The BL band structure shown in Fig. 3(a) should be contrasted with the bulk electronic band structure (Fig. 2(a)). The

![FIG. 2: GGA+U+SOC band structure of bulk ferromagnetic Fe$_3$Sn$_2$ (a) and, paramagnetic Ni$_3$Sn$_2$ (b) and Cu$_3$Sn$_2$ (c). The representative non-trivial Weyl and Dirac-like crossings have been encircled. In (a) the Fe d-orbital character are highlighted— red circle: $d_{x^2-y^2}$, green up-triangle: $d_{yz}$, cyan diamond: $d_{z^2}$, blue down-triangle: $d_{3x^2-r^2}$, yellow pentagon: $d_{3z^2-r^2}$. The corresponding non-spin-polarized projected density of states are shown for the Fe (d), Ni (e) and Cu (f) compounds.](image)
The nearly flat bands span a significant portion of the BZ and should be contrasted with the case of bulk FeSn structure consisting of kagome monolayers, where the flat band features occur at several hundreds of meV below and above $E_F$. In presence of the magnetic long range order, time-reversal symmetry is broken and these flat-ish bands acquire a finite Chern number. Constructing the maximally-localized Wannier functions (MLWFs) using the formulation of WANNIER90\cite{30}, we calculated the integrated Berry curvature over the 2D BZ. This gives a Chern number of -1 for the flat band closest to $E_F$. Thus the DFT results show that BL FeSn may stabilize a ferromagnetic Chern metal\cite{31,32} without non-quantized but large anomalous Hall response.

Effective tight binding model: Based on our DFT findings, we thus conclude, geometric confinement to BL, results in (a) quasi-2D or 2D electronic structure, (b) survival of ferromagnetic correlation, and (c) realization of low-energy bands with suppressed bandwidth—almost flat bands. The effect of fluctuations on almost flat bands are expected to be strong, opening up possibilities for stabilising novel phases. The intricate features of the low energy DFT bands near the Fermi energy (cf Fig. 3(a)) require a detailed tight-binding model accounting for the various hopping processes involved. Here instead, we construct simpler tight-binding models with the right orbital character and short range hopping that captures qualitatively the low energy DFT band structure and use them to study the effect of correlation and band properties.

To this end we introduce two related symmetry allowed models— (1) a three orbital (plus spin)/site model in which we account for the magnetisation within mean field decomposition of onsite Hubbard interactions in the ferromagnetic channel. This captures the large anomalous Hall response in the Chern metal phase (Fig. 4(b)) in a material relevant parameter regime; and, (2) an even more simplified one spin polarized orbital/site tight-binding model which captures the flatish band near the Fermi level which we use to study the possible superconductivity driven by magnetic fluctuations within a self consistent Bardeen-Cooper-Schrieffer (BCS) mean field theory. We expect that since the superconductivity arises from the instability of the Fermi surface the minimal one band model gives a valid qualitative description of such phases. The superconducting phase presents a natural instability of the Chern metal and raises a host of interesting issues regarding the fate of the system indicating that Fe$_3$Sn$_2$ can act as a rich playground of many competing conventional and unconventional correlated phases.

Our minimal tight binding modelling starts by (see SM for details; also see \cite{33}) including three Fe $d$-orbitals per-site which contribute to the electronic states within ±10 meV of the Fermi level of the DFT band structure, namely $d_{3z^2-r^2}$, $d_{x^2-y^2}$ and $d_{xy}$ (cf Fig. 3(a)). This generic symmetry allowed model allows intra and inter kagome layer first, second and third nearest neighbor hopping among the orbitals; (weak) SOC projected to the above orbitals; as well as the asymmetry of the up and down triangles (through a parameter $r$, (Fig. 5(c)) as is relevant to the material. Tuning the parameters of this model (see Tables (S4)-(S6) of the SM) we produce a representative tight-binding band structure (cf. Fig. 4(a)) in

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![FIG. 3: GGA+SOC+U band structure (a) and Fe $d$ projected spin-polarized DOS (b) of BL Fe compound in the FM state. In (a) the Fe $d$ orbital characters are denoted by point symbols with different colors, red: $d_{xy}$, green: $d_{yz}$, cyan: $d_{z^2}$, blue: $d_{3z^2-r^2}$ and yellow: $d_{z^2-y^2}$. (b) shows comparison between the BL and bulk DOS, while the inset shows the van Hove features of the two-dimensional electronic structure of the BL. (c) Band structure of BL in nonmagnetic state. (d) the Fermi surface in the non-magnetic (blue) and magnetic states (red).]
Polarised band \[34–38\]. For the rest of the work, we focus on act as a pairing glue leading to superconductivity on the spin-magnetic fluctuations. These magnetic fluctuations can then of small SOC, we expect the ferromagnetic order to be stable instability of such a metallic phase therefore involves an intri-

where the partially filled band has a topological invariant. The anomalous response can be a robust feature of possible Chern r

of the asymmetry parameter \(r\). Panel (c) shows the superconducting order parameter in the reduced one orbital spin polarised tight-binding model with nearest neighbour attractive inter-

actions mediated by ferromagnetic fluctuations. The thickness of the bands are proportional to the amplitude of the superconducting order parameter and the colors encode its phase. Panel (d) shows the topological superconductor and magnetic metal (with \(\sigma_{xy} \neq 0\)) parts of the phase diagram of the reduced tight-binding model.

semi-quantitative agreement with the DFT results (Fig. 3(a)). We note that the asymmetry parameter \(r\) is an experimentally relevant parameter which is accessible for bilayer FeSn and related materials. The tight-binding band structure presented in Fig. 4(a) which has similar characteristics as the DFT band structure for bilayer FeSn has \(r = 1.25\).

Chern Metal and Anomalous Hall effect: As suggested by our WANNIER90 Chern number calculation described above, the bands close to the Fermi level Fig. 4(a) also have non-zero Chern numbers. While it is complicated to calculate the Chern numbers of the individual bands since they cross and SOC is small, a more robust quantity is the anomalous Hall response of the resultant Chern metal. We plot the (representative) anomalous Hall conductivity (\(\sigma_{xy}\)) as a function of the asymmetry parameter \(r\) in Fig. 4(b) where other band parameters are kept fixed. The resultant finite response over a wide parameter regime \(r \in (0.7 – 1.4)\) indicates that the anomalous response can be a robust feature of possible Chern metal phase in bilayer FeSn and allied materials.

The Chern metal itself is an extremely interesting phase where the partially filled band has a topological invariant. The instability of such a metallic phase therefore involves an intricate interplay of band topology and correlations. In presence of small SOC, we expect the ferromagnetic order to be stable at finite temperatures even in the bilayer, albeit with enhanced magnetic fluctuations. These magnetic fluctuations can then act as a pairing glue leading to superconductivity on the spin-polarised band [34–38]. For the rest of the work, we focus on the possibility of realising such a magnetic fluctuation driven superconductor within a self consistent mean field theory.

Superconductivity: Both the DFT and the tight-binding model show the presence of small Fermi pockets near the \(K\) and the \(\Gamma\) points of the Brillouin zone with large density of states near the Fermi energy owing to the nearly flat spin-polarized bands involved. The nature of superconductivity that may arise in this setting can easily be explored within a mean-field theory by considering the low energy pairing instability of the above pockets. For this purpose we use a symmetry allowed effective tight binding model with one spin polarized orbital/site of the bi-layer kagome, with parameters chosen such that the DFT bands close to the Fermi-energy are well represented. Furthermore, an effective nearest neighbor attraction \(V = 2t\), where \(t\) is the nearest neighbor hopping amplitud[39], is incorporated to model an effective attractive interaction between the electrons that may get induced by the ferromagnetic-fluctuations (details in SM)[40].

In the analysis that follows we incorporate eighteen pairing order parameters corresponding to all the unique nearest neighbor bonds associated with an unit cell. These can further be classified into three groups (of six each) which do not mix under the point group symmetries of the lattice (cf. bonds of different thicknesses in Fig. 4(c)), and thus correspond to three different pair wavefunctions. Consequently, the self-consistent mean field ground state of this system exhibits superconducting order with an intricate phase pattern over the lattice. In Fig. 4(c) we show the superconducting order with the thickness of a bond being proportional its magnitude and the color of the bond encoding its phase. The maximum pairing amplitude is \(\sim 0.02t\) which would correspond to a transition temperature of \(\sim 10K\). A straightforward analysis reveals that the pairing amplitudes in Fig. 4(c) transform like a \(l_z = 1\) orbital under a rotation by \(2\pi/3\) about the centre of the hexagon of the bi-layer akin to \(\sigma_{xy} + ip_y\) topological superconductor[35]. The topological nature of superconductivity in this system is easily confirmed by computing the net Chern number of the negative energy Bogoliubov bands (see, for example, ref [41–44]). Remarkably, the topological superconducting state of the system exists for a wide range of the asymmetry parameter hinting towards its robustness and accessibility in experiments. For sufficiently small values of the asymmetry parameter superconductivity ceases to exist and one recovers a magnetic metal exhibiting anomalous Hall response. This is shown in Fig. 4(d).

Summary and Outlook: To summarise, our DFT results show that kagome intermetallic series derived from bulk \(M_2Sn_2\) (\(M=Fe, Ni, Cu\)) can host a rich interplay of band physics and correlations. To the best of our knowledge, while \(Fe_2Sn_2\) has been synthesised, the Ni and Cu counterparts are yet to be synthesised and hence provide future avenues to explore. The above interplay is most prominent in the case of Fe where dimensional confinement in the bilayer limit enhances it by stabilising a ferromagnetic metal with nearly flat bands near the Fermi level and thereby giving a Chern metal with large anomalous Hall conductivity. Instability of this Chern
metal, within a low energy tight-binding model and BCS-like mean field theory results in a topological superconductor in a material relevant parameter regime. A related instability, particularly relevant for the nearly flat band Chern metal, is a magnetic fluctuation driven fractional Chern insulator. It would be interesting to investigate the relevance of such a novel phase in the present context. All the above ingredients have close similarity with the rich physics of twisted bilayer graphene and hence experimental progress in isolating bilayer \( \text{Fe}_3\text{Sn}_2 \) and related materials may open up newer playgrounds of novel correlated physics probing the interplay of band topology, electron-electron correlations and spontaneous symmetry breaking.

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[28] In the Supplementary Material (SM) we provide further discussion on the following topics: (i) DFT computations including cleavage energy, and relaxed crystal structure, (ii) Analysis using model tight binding Hamiltonians including the computation of Hall conductivity, possible superconducting instability and its topological nature.
[29] The SOC calculations for \( \text{Fe}_3\text{Sn}_2 \) were carried out considering the magnetization axis pointed along \( x- (M_x), y- (M_y) \) or \( z- (M_z) \) directions, which showed the energy corresponding to \( M_z \) to be lower compared to \( M_x \) or \( M_y \). The results are thus shown for only \( M_z \) direction.
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[39] † must be thought of as an effective hopping amplitude which, because of correlation effects, can be much smaller than the bare hopping amplitude in the system.
[40] It has long been known that fluctuations close to a ferromagnetic transition suppress singlet pairing[36, 37] and may lead to pairing in the triplet channel. However, a detailed understanding of how the triplet pairing may get stabilized has not been achieved. Spin fluctuations (see, for example, [38]) or charge fluctuations in the presence of strong on-site repulsion (see, for example, [35]) may lead to the effective triplet pairing interaction that we include in our analysis.
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DFT COMPUTATIONAL DETAILS

The first principles electronic structure calculations have been carried out within the framework of DFT in the plane wave basis with projector augmented wave (PAW) potential[22] and generalized gradient approximation (GGA) of Perdew-Burke-Ernzerhof (PBE)[23] for exchange-correlation as implemented in Vienna *ab initio* simulation package (VASP).[24] A k-point grid of $8 \times 8 \times 2$, used to discretize the first Brillouin zone and a plane wave cut-off of 600 eV were found to be sufficient to achieve convergence of total energy with respect to k-point and energy cut-off. The energy convergence criterion was set to $10^{-8}$ eV during the energy minimization process of the self-consistent cycle.

Starting from the experimental structure, [10] full optimization of the Fe$_3$Sn$_2$ crystal structure was carried out with the force criterion of $10^{-3}$ eV/Å. The crystal structures of yet-to-be synthesized Ni$_3$Sn$_2$ and Cu$_3$Sn$_2$ were obtained by starting from the experimental structure of Fe$_3$Sn$_2$, and replacing Fe by Ni and Cu, respectively, followed by full relaxation including the cell and the atomic positions. The bilayer structures were simulated starting from the fully optimized respective bulk M$_3$Sn$_2$ (M=Fe, Ni and Cu) structures, terminated at desired layers on both side within a slab geometry. Within the periodic set-up of the calculations, the bilayer structures were separated by a sufficiently large vacuum layer of 10 Å to ensure minimal interaction between consecutive bilayers. The atomic positions in the bilayer geometries were relaxed keeping the in-plane lattice constant fixed to the value of the relaxed bulk counterparts. The electronic structure calculations for the bilayer compounds were carried out using k-point grid of $12 \times 12 \times 1$.

Due to the presence of correlated metal cations Fe, Ni and Cu, a supplemented Hubbard $U$ correction of 0.5 eV was considered at the metal sites within the framework of GGA+$U$ calculations. The choice of $U$ value of 0.5 eV was made following that in recent literature.[27] The ground state electronic structure presented in the manuscript were calculated considering both the Hubbard $U$ and the spin-orbit coupling effect using GGA+$U$+SOC method.

Cleavage Energy

Possible means to derive the 2D counterparts from the 3D layered structures are through mechanical or chemical route. An energy barrier needs to be overcome, in this respect, known as the cleavage energy. The cleavage energy ($E_{cl}$) can be defined as the energy required to generate two (top and bottom) surfaces by cleaving the bulk layered structure along the desired plane,[17, 18] $E_{cl} = 2 \times \frac{(E_{slab} - E_{bulk})}{2A}$ where $E_{slab}$ is the total energy of the cleaved system with two exposed surfaces and $E_{bulk}$ is the total energy of the same in bulk configuration, $A$ being the surface area.

Depending on the nature of the terminating surface, the atomistically thin layer limit of the studied materials can be obtained from the bulk structure in two different ways keeping intact the kagome layers, one by terminating at Sn" layer (referred as Model-I) or at Fe-Sn' layer (referred as Model-II), as shown in Fig. S1. The chemical formula of Model-I is MSn, while that of Model-II is M$_3$Sn. We calculated cleavage energies of both the models for Fe, Ni and Cu systems. For this purpose, the separations between the slabs were gradually increased in the range 0.2-10 Å and the surface area-normalized total energy differences between the slab-separated and bulk models were calculated, which show saturation when the separation exceeded 6 Å, the value at saturation being the cleavage energy. Table. S1 shows the computed values of cleavage energies for all three compounds both in Model-I and Model-II.

Our calculation of the cleavage energies showed that the termination at the Sn" layer to be somewhat favorable over that at Fe-Sn' layer, as found in recent study.[16] In the text we thus focused only on Model-I.
Crystal Structures

The fully relaxed crystals of the bulk Fe$_3$Sn$_2$, Ni$_3$Sn$_2$ and Cu$_3$Sn$_2$ and the corresponding bilayers are tabulated in the Table. S2 and Table. S3, respectively. In the bulk geometry, the metal ions form bilayers of kagome networks, consisting of big and small equilateral triangles. Sn’ atoms occupy the position close to the center of the hexagons in the kagome planes, while the kagome bilayers are separated by Sn” layers. The optimized geometries show that for Ni and Cu compounds, the Sn’ atoms are pushed further towards the direction of kagome plane in case Ni and away in case of Cu, compared to that of Fe. The difference in metal-metal bond length, $d_{M-M} \pm 0.03$ Å between the two triangles of the metal cations, was found to decrease with increase in atomic number from Fe to Cu. The difference between big and small triangles were found to be diminished for Ni compounds, with metal-metal distances of 2.73 Å and 2.55 Å, compared to that of Fe compounds (2.78 Å and 2.54 Å) (cf. Table. S2). The difference in $d_{M-M}$ between big and small triangles was found to be marginally effected in moving from 3D to bilayer geometries.

| Fe$_3$Sn$_2$, space group=R-3m (166), a=b=5.328 Å,c=19.791 Å, $\beta=120^\circ$ |
|----------------------------------|----------------|
| Sites Wyckoff x y z d$^{M-M}$ (Å) |
| Fe h 0.17416 -0.17416 0.21958 2.78,2.54 |
| Sn’ c 0.0 0.0 0.10536 |
| Sn” c 0.0 0.0 0.33054 |

| Ni$_3$Sn$_2$, space group=R-3m (166), a=5.283 Å,c=19.715 Å, $\beta=120^\circ$ |
|----------------------------------|----------------|
| Sites Wyckoff x y z d$^{M-M}$ (Å) |
| Ni h 0.17239 -0.17239 0.22493 2.73,2.55 |
| Sn’ c 0.0 0.0 0.11595 |
| Sn” c 0.0 0.0 0.33005 |

| Cu$_3$Sn$_2$, space group=R-3m (166), a=5.562 Å,c=19.268 Å, $\beta=120^\circ$ |
|----------------------------------|----------------|
| Sites Wyckoff x y z d$^{M-M}$ (Å) |
| Cu h 0.16735 -0.16735 0.21831 2.79,2.77 |
| Sn’ c 0.0 0.0 0.09249 |
| Sn” c 0.0 0.0 0.33218 |

TABLE II: Optimized geometry of bulk Fe$_3$Sn$_2$, Ni$_3$Sn$_2$ and Cu$_3$Sn$_2$

ANALYSIS USING MODEL TIGHT BINDING HAMILTONIAN

To model the low energy band structure of bilayer Fe$_3$Sn$_2$ (Fig. 1(c) in the main text), we start by considering the following Fe spinful orbitals/site \{d$_{x^2-y^2}^{Fe}$, d$_{x'y'}^{Fe}$\}, following the DFT results. To take full advantage of the lattice symmetries we define a site dependent orientation of these orbitals similar to Ref 33. Note that this local rotation of the orbitals enforces certain constraints on the relative signs of the inter-orbital hopping amplitudes between the orbitals at different sites which we have taken into account.

The resultant tight-binding model has the generic form

$$H = \sum_{\alpha,\beta} H_{\alpha\beta} = \sum_{\alpha,\beta} \left[ (1 - \delta_{\alpha\beta}) (H_{soc}^{\alpha\beta} + H_{nn}^{\alpha\beta}) + \delta_{\alpha\beta} (H_{soc}^{\alpha\beta} + H_{nn}^{\alpha\beta} + H_{nnn}^{\alpha\beta}) \right]$$

where $\alpha$ and $\beta$ denote the orbitals; inter-orbital and intra-orbital terms have been made explicit. In eq. (1) $H_{soc}^{\alpha\beta}$ is the onsite term originating from the onsite energy (the kagome crystal field splits the $d_{x^2-y^2}$ orbital from the $d_{x'y'}$ orbital; cf Ref. 33) and the background magnetization (arising from a mean field treat of the onsite interaction term $U_{n\uparrow}n_{\downarrow} = \frac{U}{2} [n_{\uparrow} + n_{\downarrow} - \frac{1}{3}S^2] \simeq \frac{U}{2} [n_{\uparrow} + n_{\downarrow} - S^2_{\uparrow}]$)

which appears as a multiplicativ factor with nearest neighbor hopping amplitudes between an orbital inside the unit cell and another outside the unit cell. $H_{soc}^{\alpha\beta}$ and $H_{nn}^{\alpha\beta}$ denote the next-to-nearest-neighbor and next-to-next-nearest-neighbor hopping terms. The SOC, though small, plays an important role. It is obtained by projecting the atomic spin-orbit coupling to the above manifold of orbitals. The projected Hamiltonian has the following second quantized form

$$H_{soc}^{\alpha\beta} = i\lambda_{xy} \sum_{i,s,\gamma} \left[ c_{i3s\gamma}^{\dagger} c_{i2s'\gamma'} + c_{i3s\gamma}^{\dagger} c_{i2s'\gamma'} - c_{i2s\gamma}^{\dagger} c_{i3s'\gamma'} + c_{i2s\gamma}^{\dagger} c_{i3s'\gamma'} \right]$$
where the subscripts $i$, $\chi$, $s$, $\gamma$ and $\sigma$ indicate lattice site, orbital ($\chi \in \{d_{z^2-r^2}, \tilde{d}_{x^2-y^2}, \tilde{d}_{xy}\} \equiv \{1, 2, 3\}$), site index within a unit cell, layer number and spin, respectively. The relevant parameters in our model and their meanings are shown schematically in Fig. 6 and enumerated in Tables (IV)-(VI).

### Computation of DC Hall conductivity

The real part of DC Hall conductivity, $\sigma_{xy}$, within linear response theory is

$$\sigma_{xy} = \sum_{n \neq 0} \frac{\langle 0|J_y|n\rangle \langle n|J_x|0\rangle - \langle 0|J_x|n\rangle \langle n|J_y|0\rangle}{(E_n - E_0)^2}$$  \hspace{1cm} (3)

where $J_\mu$ is the total current operator and $E_n(0)$ is the energy of the eigenstate $|n\rangle$ ($0 \equiv$ ground state) of the system. The hopping Hamiltonian can easily be diagonalized to compute the Hall conductivity for our TB model. The result is plotted in Fig. 4(b) of the main text as a function of the anisotropy parameter $r$.

### Superconductivity

As discussed in the main text, to capture the low energy superconducting instability of the Fermi surface we use a one spin-polarised orbital/site TB model given by Eq. 1 where the symmetry allowed effective SOC is given by $\tilde{H}_{soc} = \sum_{(i,j)} i\lambda \nu_{ij} c_{i\uparrow}^{\dagger} c_{j\uparrow}$, where $\lambda$ is the effective spin-orbit coupling parameter and $\nu_{ij} = +1(-1)$ if $j \rightarrow i$ is anti-clockwise (clockwise) around the triangle containing the sites $i$ and $j$ (cf ref 33). The parameters for our reduced model, presented in Table VII, are chosen so that the qualitative features of the DFT bands close to the Fermi energy are adequately represented (see Fig. 7).

We now introduce an effective attractive interaction ($V = 2t$) on the nearest neighbor bonds, possibly arising because of spin-fluctuations, and solve for the most generic mean-field superconducting state which does not break lattice translation symmetry. This state has eighteen variational parameters (pairing amplitudes) shown in Fig. 8. The mean field Hamiltonian thus obtained, $\tilde{H}_{SC}$, and can be put in the Nambu form

$$\tilde{H}_{SC} = \sum_{k>0} \psi_k^\dagger H_k \psi_k \quad \text{where} \quad \psi_k \equiv \left[ c_{k1\uparrow}^{\dagger}, c_{k2\uparrow}^{\dagger}, c_{k3\uparrow}^{\dagger}, c_{k12\uparrow}^{\dagger}, c_{k23\uparrow}^{\dagger}, c_{k31\uparrow}^{\dagger}, c_{-k22\uparrow}^{\dagger}, c_{-k32\uparrow}^{\dagger}, c_{-k12\uparrow}^{\dagger}, c_{-k31\uparrow}^{\dagger}, c_{-k11\uparrow}^{\dagger} \right]$$  \hspace{1cm} (4)

and $\{c_{k\chi\gamma\sigma}\}$ are the Fourier components of $c_{i\chi s\gamma\sigma}$ in eq. (2) with the orbital index $\chi$ dropped. $\tilde{H}_{SC}$ is then diagonalized using Bogoliubov transformation and all the pairing amplitudes are determined self-consistently. The dominant pairing amplitudes are shown in Fig. 9 as a function of the asymmetry parameter.

### Computing Chern number for the Bogoliubov bands

With the self consistent pairing amplitudes, we calculate the Chern numbers of the negative energy Bogoliubov bands and their sum can be used to characterize the topological nature of the mean field superconducting state of our system\[42–44\. It turns out this sum is odd for the SC that our system hosts, establishing its non-trivial topological nature (see Fig 4(d) of the main text).
FIG. 6: **Hopping structure:** (Color online) The hopping structure of each orbital species out-of-plane and in-plane are shown in the left and right diagrams, respectively. The unit cell is marked with indices in the left. **Left schematic** There is one out-of-plane nearest neighbor (nn) hopping \( t_{x} \) and it operates only within a unit cell (solid grey lines). There are two kinds of out-of-plane next nearest neighbor (nnn) hoppings, \( t_{L1X} \) which operates within a unit cell (dashed grey lines) and \( t'_{L1X} \) which operates between unit cells (dotted grey lines). **Right schematic** The nn hopping \( t_{x} \) is shown by solid orange lines for the lower layer. There in only one variety of nnn hopping \( t_{1X} \) which has been shown by dotted green lines. There are two varieties of next to next nearest neighbor (nnnn) hoppings. The first one operates along the solid orange lines \( t_{2X} \) (dashed red lines) and the second one passes through the centre of the hexagons \( t'_{2X} \) (dashed blue lines). The hopping structure in the upper layer can be obtained by rotating the lower plane by 180° about the centre of any up triangle.

| parameters | description | value          |
|------------|-------------|----------------|
| \( a_1 \)  | first lattice vector | \((1, 0)\) |
| \( a_2 \)  | second lattice vector | \((1/2, \sqrt{3}/2)\) |
| \( R_1^\perp \) | first reciprocal lattice vector | \(2\pi(1, -1/\sqrt{3})\) |
| \( R_2^\perp \) | second reciprocal lattice vector | \(2\pi(0, 2/\sqrt{3})\) |

**TABLE IV: Lattice parameters:** Lattice vectors and reciprocal lattice vectors.

| parameters | description | \( \chi = 1 \) | \( \chi = 2 \) | \( \chi = 3 \) |
|------------|-------------|----------------|----------------|----------------|
| \( t_x \)  | nn in-plane hopping between Fe sites inside the unit cell | -0.07 | -0.405 | 0.185 |
| \( r_x \)  | ratio between the nn hopping parameters outside the unit cell to those inside the unit cell | 1.25 | 1.25 | 1.25 |
| \( t_{L1X} \) | nn out-of-plane hopping between Fe sites | 0.06 | -0.185 | 0.045 |
| \( t_{L1X} \) | nn in-plane hopping between Fe sites | -0.03 | 0.075 | -0.135 |
| \( t'_{L1X} \) | nn out-of-plane hopping between Fe sites within a unit cell | 0.02 | 0.06 | -0.01 |
| \( t_{L1X} \) | nnn out-of-plane hopping between Fe sites connecting different unit cells | 0.03 | -0.09 | -0.08 |
| \( t_{2X} \)  | one variety of nnnn in-plane hopping between Fe sites | -0.01 | -0.03 | 0.075 |
| \( t'_{2X} \) | another variant of nnnn in-plane hopping between Fe sites | 0.01 | 0.105 | -0.045 |
| \( h_x \)  | background magnetization | 1.35 | 1.35 | 1.35 |
| \( \epsilon_x \) | onsite energy of the orbitals | 0.69 | 1.82 | 1.82 |

**TABLE V: Intra-orbital parameters:** Parameters which connect orbitals of the same flavor (\( \chi \)) including onsite and hopping terms (for details see Fig. 6). \( \chi \in \{d_{3z^2-r^2}, d_{2-y^2}, d_{xy}\} \equiv \{1, 2, 3\} \).

| parameters | description | value |
|------------|-------------|-------|
| \( t_{12} \)  | nn in-plane hopping between \( d_{3z^2-r^2} \) and \( d_{2-y^2} \) orbitals at Fe sites inside the unit cell | -0.04 |
| \( r_{12} \)  | ratio between the nn hopping between \( d_{3z^2-r^2} \) and \( d_{2-y^2} \) orbitals outside the unit cell to those inside the unit cell | 1.25 |
| \( t_{13} \)  | nn in-plane hopping between \( d_{3z^2-r^2} \) and \( d_{xy} \) orbitals at Fe sites inside the unit cell | 0.05 |
| \( r_{13} \)  | ratio between the nn hopping between \( d_{3z^2-r^2} \) and \( d_{xy} \) orbitals outside the unit cell to those inside the unit cell | 1.25 |
| \( t_{23} \)  | nn in-plane hopping between \( d_{2-y^2} \) and \( d_{xy} \) orbitals at Fe sites inside the unit cell | 0.14 |
| \( r_{23} \)  | ratio between the nn hopping between \( d_{2-y^2} \) and \( d_{xy} \) orbitals outside the unit cell to those inside the unit cell | 1.25 |
| \( \lambda_{23} \) | strength of spin-orbit coupling which operates only between \( d_{2-y^2} \) and \( d_{xy} \) orbitals | 0.11 |

**TABLE VI: Inter-orbital parameters:** Parameters which connect orbitals of different flavor, including the spin-orbit coupling term.
The amplitudes of the order parameters have been plotted on the left axis while their phases have been plotted on the right axis. Every other parameter is the same as in Table VII. Clearly, the results are robust to the different discretizations.}

![Diagram](https://via.placeholder.com/150)

**FIG. 7:** (Color online) The tight binding bands obtained for parameters presented in Table (VII).

**TABLE VII:** Parameters for reduced TB model: Parameters of our one-orbital / site model which capture important features of the DFT bands close to $\varepsilon_F$.

| parameter | $t$ | $r$ | $t_{+1}$ | $t_1$ | $t_{+1}$ | $t_{-1}$ | $t_{+2}$ | $t_{-2}$ | $h$ | $\epsilon$ | $\lambda$ |
|-----------|-----|-----|----------|-------|----------|----------|----------|----------|-----|----------|--------|
| values    | 0.13| 1.35| 0.06     | -0.1  | 0.02     | 0.0      | 0.05     | 0.01     | 1.35 | 1.67     | 0.01   |

![Diagram](https://via.placeholder.com/150)

**FIG. 8:** (Color online) The eighteen pairing amplitudes associated with an unit cell (marked by indices in the left side of the figure) are shown. The arrows on the bonds show the ordering of the fermionic operators used in the definition of the pairing amplitudes. For example, $\Delta_{\perp,1} = \frac{V}{N} \sum_i c_{i11}^\dagger c_{i12}^\dagger$.

![Diagram](https://via.placeholder.com/150)

**FIG. 9:** (Color online) The out-of-plane SC order parameters (dominant bonds in Fig 4(c) of main text, i.e., $\Delta_{\perp,1}$, $\Delta_{\perp,2}$, $\Delta_{\perp,3}$, $\Delta_{\perp,4}$, $\Delta_{\perp,5}$, $\Delta_{\perp,6}$) have been plotted as a function of $r$. The amplitudes of the order parameters have been plotted on the left axis while their phases have been plotted on the right axis. Every other parameter is the same as in Table VII. Clearly, the results are robust to the different discretizations ($N_k \times N_k$) of the BZ used.