# Atomic layer epitaxy of kagome magnet Fe<sub>3</sub>Sn<sub>2</sub> and Sn-modulated heterostructures

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## Atomic layer epitaxy of kagome magnet Fe<sub>3</sub>Sn<sub>2</sub> and Sn-modulated heterostructures

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#### ABSTRACT

Magnetic materials with kagome crystal structure exhibit rich physics, such as frustrated magnetism, skyrmion formation, topological flat bands, and Dirac/Weyl points. Until recently, most studies on kagome magnets have been performed on bulk crystals or polycrystalline films. Here, we report the atomic layer molecular beam epitaxy synthesis of high-quality thin films of topological kagome magnet  $Fe_3Sn_2$ . The structural and magnetic characterization of  $Fe_3Sn_2$  on epitaxial Pt(111) identifies highly ordered films with c-plane orientation and an inplane magnetic easy axis. Studies on the local magnetic structure by anomalous Nernst effect imaging reveal in-plane oriented micrometer size domains. Superlattice structures consisting of  $Fe_3Sn_2$  and  $Fe_3Sn$  are also synthesized by atomic layer molecular beam epitaxy the ability to modulate the sample structure at the atomic level. The realization of high-quality films by atomic layer molecular beam epitaxy opens the door to explore the rich physics of this system and investigate novel spintronic phenomena by interfacing  $Fe_3Sn_2$  with other materials.

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In recent years, studies on magnetic topological materials with kagome lattices have become one of the hottest frontiers of condensed matter research, owing to their exotic physical properties in both real space and momentum space.<sup>1,2</sup> In momentum space, angle-resolved photoemission spectroscopy (ARPES) experiments on  $Mn_3Sn$ ,  $Fe_3Sn_2$ , FeSn, and  $CoSn^{3-6}$  show that kagome lattices give rise to Dirac cones and flat bands that are topologically protected and are of particular interest. In addition, scanning tunneling spectroscopy finds evidence for topological flat bands as a sharp peak in the local density of states.<sup>7</sup> These topologically nontrivial features result in signatures of anomalous transport (e.g., chiral anomaly) in magnetotransport experiments.<sup>3,8</sup> Furthermore, it is theoretically predicted that the band structures of the kagome topological magnets can be controlled by tuning their magnetic structures.<sup>1,3</sup> In real space, the kagome topological magnets have layered structures with spins occupying corner-sharing triangular lattices, which leads

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to geometrical spin frustration.<sup>9,10</sup> A surprisingly large anomalous Hall effect (AHE) and magneto-optic Kerr effect (MOKE) have been reported in noncollinear antiferromagnet Mn<sub>3</sub>Sn, even with vanishingly small net magnetization.<sup>10,11</sup> Skyrmion spin textures have been observed in ferromagnetic Fe<sub>3</sub>Sn<sub>2</sub> resulting from the competition of exchange, dipolar, and Zeeman energies.<sup>12,13</sup> However, most of the studies on the kagome magnets have been done on bulk materials<sup>4,5,9–15</sup> with a few papers reporting the growth and characterization of epitaxial films.<sup>16-20</sup> Looking forward, the heterostructures consisting of kagome magnets will be interesting for both fundamental research and applications, owing to the possibility of tuning the magnetic and topological properties via interface interactions, epitaxial strain, and quantum confinement. However, all the reported studies on epitaxial Fe<sub>3</sub>Sn<sub>2</sub> thin films have been focusing on high temperature growth so far, which may not allow for the formation of well-defined heterostructures due to interdiffusion at elevated temperatures. Therefore, lower temperature growth of  $Fe_3Sn_2$  is desired for the future development of heterostructures and superlattices based on kagome magnets.

In this paper, we report the atomic layer molecular beam epitaxy (AL-MBE) growth of high-quality Fe<sub>3</sub>Sn<sub>2</sub> thin films on Pt(111)/Al<sub>2</sub>O<sub>3</sub>(0001) substrates at lower temperatures. By sequentially depositing Fe<sub>3</sub>Sn kagome layers and Sn<sub>2</sub> layers [see Fig. 1(a)], we can control the sample structure at the atomic level. The crystalline structure of our Fe<sub>3</sub>Sn<sub>2</sub> sample is confirmed by a combination of in situ reflection high energy electron diffraction (RHEED), x-ray diffraction (XRD), and transmission electron microscopy (TEM). Energy-dispersive x-ray spectroscopy (EDX) shows sharp interfaces for low temperature growth. The magnetic properties of Fe<sub>3</sub>Sn<sub>2</sub> are investigated by using MOKE, superconducting quantum interference device (SQUID), and anomalous Nernst effect (ANE). Using a microscopy technique based on ANE, we successfully image the in-plane oriented domain structure of the epitaxial Fe<sub>3</sub>Sn<sub>2</sub> films and investigate the magnetization reversal as a function of the applied field. We further utilize AL-MBE to precisely control the stacking sequences of Fe<sub>3</sub>Sn and Sn<sub>2</sub> atomic layers, making superlattices with modulation of Sn<sub>2</sub> layers, and confirm their structures by TEM and EDX. This demonstrates the potential of using AL-MBE to generate designer materials consisting of kagome layers (Mn<sub>3</sub>Sn, Fe<sub>3</sub>Sn, Co<sub>3</sub>Sn, etc.) and Sn<sub>2</sub> spacer layers with precision control of sample structures at the atomic level.

Fe<sub>3</sub>Sn<sub>2</sub> is a ferromagnet with a high Curie temperature,  $T_C = 670 \text{ K}^{21}$ , and saturation magnetization of  $1.9 \mu_B$  per Fe at low temperature.<sup>4</sup> Figure 1(a) shows the crystal structure of Fe<sub>3</sub>Sn<sub>2</sub> (space group R3m, with lattice constants a = 5.338 Å and c = 19.789Å<sup>21</sup>), which consists of Fe<sub>3</sub>Sn kagome layers and Sn<sub>2</sub> spacer layers. In each Fe<sub>3</sub>Sn monolayer, the Fe atoms form corner-sharing equilateral triangles surrounding hexagons, with Sn atoms sitting in the center of the hexagons. The alternating sequence of one Sn<sub>2</sub> monolayer with honeycomb lattice and two Fe<sub>3</sub>Sn kagome layers produces the layered crystal structure of Fe<sub>3</sub>Sn<sub>2</sub>.

Based on this layered structure, we synthesized Fe<sub>3</sub>Sn<sub>2</sub> thin films on top of epitaxial Pt(111) buffer layers on Al<sub>2</sub>O<sub>3</sub>(0001) substrates by AL-MBE. The epitaxial growth was performed in an MBE chamber with a base pressure of  $4 \times 10^{-10}$  Torr. Films were deposited on Al<sub>2</sub>O<sub>3</sub>(0001) substrates (MTI Corporation) prepared by annealing in air at 1000 °C for 3 h followed by annealing in ultrahigh vacuum (UHV) at 500 °C for 30 min. A 5 nm Pt(111) buffer laver was deposited from an e-beam evaporator (Pt: 99.99%, Kurt J. Lesker) onto the Al<sub>2</sub>O<sub>3</sub> (0001) substrate by growing the first 0.6 nm at 440 °C and the rest 4.4 nm while cooling down from 140 to 80 °C. The Pt buffer layer was post-annealed at 300 °C to improve the crystallinity and surface roughness. The Fe<sub>3</sub>Sn<sub>2</sub> layer was grown on Pt(111) at 100 °C by using the following AL-MBE sequence: deposit two atomic layers of Fe<sub>3</sub>Sn with a Fe:Sn flux ratio of 3:1, deposit one atomic layer of Sn<sub>2</sub> with the growth time same as two Fe<sub>3</sub>Sn layers, and then repeat. The Fe and Sn fluxes were generated from Knudsen cells (Fe: 99.99%, Alfa Aesar; Sn: 99.998%, Alfa Aesar), and the growth rates were determined by using a quartz deposition monitor that was calibrated by x-ray reflectometry. Typical growth rates were ~0.85 Å/min, ~0.67 Å/min, and ~0.45 Å/min for Fe, Sn, and Pt, respectively. To protect the sample from oxidation, a 5 nm CaF<sub>2</sub> capping layer was deposited on top of the Fe<sub>3</sub>Sn<sub>2</sub>.



FIG. 1. Material structure and growth. (a) Left: Top view of an individual Fe<sub>3</sub>Sn layer with kagome structure (top) and Sn<sub>2</sub> layer with honeycomb structure (bottom). Right: Side view of the crystal structure of Fe<sub>3</sub>Sn<sub>2</sub> consisting of alternating stacking of two Fe<sub>3</sub>Sn kagome layers and one Sn<sub>2</sub> layer. (b) RHEED patterns for the Al<sub>2</sub>O<sub>3</sub>(0001) substrate, 5 nm Pt film, and 20 nm Fe<sub>3</sub>Sn<sub>2</sub> film measured with the beam along [1100] (left column) and [1120] (right column) directions of the substrate. (c) Oscillations in the normalized RHEED intensity as a function of time. The RHEED intensity is measured within the red box and normalized by the intensity of the whole image as background.

RHEED patterns were measured during growth to characterize the epitaxial growth and determine the in-plane lattice constants. Figure 1(b) shows the RHEED patterns for the Al<sub>2</sub>O<sub>3</sub>(0001) substrate (top row), the 5 nm Pt buffer layer (middle row), and the Fe<sub>3</sub>Sn<sub>2</sub> layer after 20 nm of growth (bottom row). The left and right columns show the patterns taken for the beam along the [1100] and [1120] directions of the substrate, respectively. With in-plane rotation of the sample, RHEED exhibits the same pattern every 60° (i.e., sixfold rotation symmetry) with the patterns alternating between [1100]-type and [1120]-type every 30°. For the in-plane epitaxial alignment, the Pt(111) and Fe<sub>3</sub>Sn<sub>2</sub>(0001) surface unit cells are aligned with each other and rotated 30° with respect to the Al<sub>2</sub>O<sub>3</sub>(0001) substrate, as  $a_{Al_2O_3} \approx (\sqrt{3}/2)2a_{Pt} \approx (\sqrt{3}/2)a_{Fe_3Sn_2}$ (bulk values for in-plane lattice constant are  $a_{Al_2O_3} = 4.759$  Å,  $2a_{Pt}$ = 5.549 Å, and  $a_{Fe_3Sn_2} = 5.338$  Å).

The streaky patterns observed during  $Fe_3Sn_2$  growth signify diffraction from a two-dimensional surface. In addition, we observe oscillations [Fig. 1(c)] in the normalized RHEED intensity where the

maxima occurs for the  $Fe_3Sn$  termination and the minima occurs for the  $Sn_2$  termination. The normalization is performed by dividing the intensity of the background and is helpful for canceling variations in the incident beam intensity and background lighting. Except for the change in the RHEED intensity, we did not observe any other significant difference in the RHEED patterns between  $Sn_2$  and  $Fe_3Sn$  terminations. Nevertheless, the presence of the RHEED oscillations in atomic layer MBE confirms the modulation of the surface termination during growth.

Films grown by this method were studied with XRD using the Cu K $\alpha$  line (wavelength 1.5406 Å) to analyze their crystal structure. A representative  $2\theta$ - $\omega$  scan of a 20 nm film grown at 100 °C is shown in Fig. 2(a) and includes the Fe<sub>3</sub>Sn<sub>2</sub> (0009) peak with several Laue oscillations, indicating a smooth film. The out-of-plane lattice parameter extracted from the analysis of this scan is 19.85 Å, which agrees well with previous reports of 19.789 Å.<sup>21</sup> A peak from the 5 nm Pt(111) buffer also shows the Laue oscillations with larger angular period due to smaller thickness of the Pt layer in



**FIG. 2.** Material characterization. (a)  $2\theta$ - $\omega$  scan of a 20 nm Fe<sub>3</sub>Sn<sub>2</sub> film grown on c-sapphire. (b) AFM scan over a  $10 \times 10 \ \mu\text{m}^2$  area on the surface of a 20 nm Fe<sub>3</sub>Sn<sub>2</sub> film (rms roughness 0.362 nm). (c) Cross-sectional EDX-STEM chemical map of a 20 nm Fe<sub>3</sub>Sn<sub>2</sub> sample grown at 100 °C. (d) Atomic resolution HAADF-STEM imaging of a 20 nm Fe<sub>3</sub>Sn<sub>2</sub> sample grown at 100 °C. (e) Atomic resolution HAADF-STEM imaging of a 20 nm Fe<sub>3</sub>Sn<sub>2</sub> sample grown at 100 °C. (f) Atomic resolution HAADF-STEM imaging of a 20 nm Fe<sub>3</sub>Sn<sub>2</sub> sample grown at 100 °C. (f) Atomic resolution HAADF-STEM imaging of a 20 nm Fe<sub>3</sub>Sn<sub>2</sub> sample grown at 100 °C. (f) Atomic resolution HAADF-STEM imaging of a 20 nm Fe<sub>3</sub>Sn<sub>2</sub> sample grown at 100 °C. (f) Atomic resolution HAADF-STEM imaging of a 20 nm Fe<sub>3</sub>Sn<sub>2</sub> sample grown at 100 °C. (f) Atomic resolution HAADF-STEM imaging of a 20 nm Fe<sub>3</sub>Sn<sub>2</sub> sample grown at 100 °C. (f) Atomic resolution HAADF-STEM imaging of a 20 nm Fe<sub>3</sub>Sn<sub>2</sub> sample grown at 100 °C. (f) Atomic resolution HAADF-STEM imaging of a 20 nm Fe<sub>3</sub>Sn<sub>2</sub> sample grown at 100 °C. (f) Atomic resolution HAADF-STEM imaging of a 20 nm Fe<sub>3</sub>Sn<sub>2</sub> sample grown at 100 °C. (f) Atomic resolution HAADF-STEM imaging of a 20 nm Fe<sub>3</sub>Sn<sub>2</sub> sample grown at 100 °C. (f) Atomic resolution HAADF-STEM imaging of a 20 nm Fe<sub>3</sub>Sn<sub>2</sub> sample grown at 100 °C. (f) Atomic resolution HAADF-STEM imaging of a 20 nm Fe<sub>3</sub>Sn<sub>2</sub> sample grown at 100 °C. (f) Atomic resolution HAADF-STEM imaging f) Atomic

comparison with Fe<sub>3</sub>Sn<sub>2</sub> layer (Pt Laue peaks at ~34.5°, ~36.7°, and ~43.5°), demonstrating the high quality of the buffer layer. At larger  $2\theta$  angles, we observe Fe<sub>3</sub>Sn<sub>2</sub> (00018) and Pt(222) peaks, with no additional peaks that could be attributed to the impurity phases (see the supplementary material, Sec. 2, for full range scans and rocking curve data).

To characterize the surface topography of the sample, we performed atomic force microscope (AFM) measurements on uncapped 20 nm Fe<sub>3</sub>Sn<sub>2</sub> films. Figure 2(b) shows a typical  $10 \times 10 \ \mu\text{m}^2$  scan of a 20 nm Fe<sub>3</sub>Sn<sub>2</sub> sample grown at 100 °C. The AFM image indicates that the sample has a flat surface, with a root-mean-square (rms) roughness of 0.362 nm.

An important factor for material synthesis is the growth temperature. To optimize the growth temperature, we performed AFM and XRD measurements on a series of samples grown at different temperatures ranging from room temperature to 200 °C. The AFM and XRD results are shown in the supplementary material, Secs. 1 and 2, respectively. We conclude that 100 °C is the optimized growth temperature as it gives the best AFM roughness and sharp XRD peaks.

The epitaxial quality of the 20 nm Fe<sub>3</sub>Sn<sub>2</sub> sample grown at 100 °C was examined by using a probe-corrected Themis Z S/TEM at 200 kV. Figures 2(c) and 2(d) show the energy-dispersive x-ray (EDX) chemical map and cross-sectional scanning transmission electron microscopy (STEM) image, revealing a clear interface between the Pt buffer and Fe<sub>3</sub>Sn<sub>2</sub> thin film. The stoichiometry of Fe<sub>3</sub>Sn<sub>2</sub> thin films was confirmed by electron energy loss spectroscopy (EELS), which gives an atomic ratio of Fe:Sn  $\approx$  1.5 (see the supplementary material, Sec. 3, for details). To identify the crystalline quality of the Fe<sub>3</sub>Sn<sub>2</sub> thin films, atomic resolution high-angle annular dark-field (HAADF) STEM images of the Fe<sub>3</sub>Sn<sub>2</sub>/Pt interface were acquired along the  $Fe_3Sn_2$  [1120] direction [see Fig. 2(d)]. Since the contrast in HAADF STEM is approximately proportional to the square of the atomic number, the Sn atoms in the Sn<sub>2</sub> atomic layers appear as the brightest spots, while the atoms in Fe<sub>3</sub>Sn kagome layers are dimmer (the atomic numbers of Sn and Fe are 50 and 26, respectively). The alternating sequence of one  $Sn_2$  monolayer and two Fe<sub>3</sub>Sn kagome layers shows a highly crystalline film with the expected Fe<sub>3</sub>Sn<sub>2</sub> phase, although some stacking faults are observed.

To investigate the in-plane and out-of-plane magnetic properties of the Fe<sub>3</sub>Sn<sub>2</sub> films, we measured the longitudinal and polar MOKE hysteresis loops. The samples were probed using a linearly polarized He–Ne laser (633 nm wavelength, ~100  $\mu$ W power, and ~100  $\mu$ m spot size) and a polarizing beamsplitter, photodiode bridge, and lock-in amplifier (463 Hz intensity modulation) to detect the Kerr rotation. The laser beam had a  $\sim 45^{\circ}$  angle of incidence for longitudinal MOKE and normal incidence for polar MOKE. Figure 3(a) shows a representative longitudinal hysteresis loop (red curve) measured on a 20 nm thick Fe<sub>3</sub>Sn<sub>2</sub> sample. The square hysteresis loop with a coercivity of 2.4 mT indicates ferromagnetic order with in-plane magnetization. In contrast, the polar hysteresis loop (blue curve) shows a small Kerr rotation with slight variation with the out-of-plane magnetic field. Together, the longitudinal and polar MOKE loops show that the Fe<sub>3</sub>Sn<sub>2</sub> samples have an easy-plane magnetic anisotropy. This agrees with previous studies of Fe<sub>3</sub>Sn<sub>2</sub> films grown by sputter deposition<sup>19</sup> and in bulk crystals thinned to below ~100 nm.<sup>22</sup> In addition, Fe<sub>3</sub>Sn<sub>2</sub> thin films have very weak magnetic anisotropy within the sample plane, which is shown by our longitudinal MOKE data as a function of different in-plane directions (see the supplementary material, Sec. 4).

We also performed SQUID magnetometry measurements on our Fe<sub>3</sub>Sn<sub>2</sub> films (see the supplementary material, Sec. 5, for SQUID measurement details). Figure 3(b) shows hysteresis loops of a 20 nm thick Fe<sub>3</sub>Sn<sub>2</sub> sample measured with in-plane (red curve) and outof-plane (blue curve) magnetic fields. The in-plane hysteresis loop exhibits a sharp switching behavior, while the out-of-plane hysteresis loop exhibits almost linear behavior within ±1 T and saturates at ~1 T, suggesting that our Fe<sub>3</sub>Sn<sub>2</sub> samples have easy-plane anisotropy. Furthermore, our SQUID results give a saturation magnetization  $M_s = 1.9 \mu_B/\text{Fe}$  (630 kA/m), which is consistent with previous studies.<sup>14,19</sup>



FIG. 3. Magnetic properties of Fe<sub>3</sub>Sn<sub>2</sub> films. (a) Longitudinal (red) and polar (blue) MOKE hysteresis loops of a 20 nm Fe<sub>3</sub>Sn<sub>2</sub> film. (b) Magnetic hysteresis loops of a 20 nm Fe<sub>3</sub>Sn<sub>2</sub> film measured using SQUID magnetometry with in-plane (red) and out-of-plane (blue) geometries.

The magnetic domain structures of  $Fe_3Sn_2$  films are of interest due to the observation of skyrmions in bulk  $Fe_3Sn_2$ , but have not yet been studied in thin films. Longitudinal MOKE microscopy with oblique angle incidence can detect the in-plane magnetization and, therefore, determine the in-plane domain structure of our  $Fe_3Sn_2$  films. However, in this paper, we choose to use thermal gradient microscopy (TGM)<sup>23–25</sup> over longitudinal MOKE to image the domain structure because we found that it has a better signal-to-noise ratio in our experimental setup.

TGM is based on moving a laser spot over the sample surface and recording a voltage induced by the local laser heating. The thermal gradient generated in the out-of-plane direction Z and a component of magnetization in the X direction give rise to the anomalous Nernst effect, which is detected as a voltage along the Y direction,  $V_{ANE} \sim [\nabla T \times \mathbf{M}]$  [see Fig. 4(a)].

For the ANE imaging, we fabricated  $10 \,\mu$ m wide Hall bar devices by a combination of photolithography and argon ion milling [Fig. 4(b)]. The laser excitation for the thermal gradient was produced by a frequency-doubled (BaB<sub>2</sub>O<sub>4</sub> crystal) mode-locked Ti:sapphire laser for a wavelength of 400 nm. The laser beam with 0.7 mW power was focused by a 50× objective lens (NA of 0.6) to a spot size of 0.9  $\mu$ m, and a fast steering mirror in the 4f alignment scheme was used for scanning the laser spot over the sample surface. The intensity of the beam was modulated at a frequency of 120 kHz and the generated ANE voltage was detected using a lock-in amplifier.

We first utilized the ANE microscope to measure a detailed hysteresis loop at a fixed position. As shown in Fig. 4(c) for the magnetic

field along the X direction, the hysteresis loop shows a gradual reversal followed by magnetization switching with a coercivity of 2.3 mT. This has a similar coercivity but more gradual initial reversal than the in-plane hysteresis loops obtained by MOKE [Fig. 3(a)]. The origin of the different hysteresis properties is revealed by imaging the magnetic domain structure of Fe<sub>3</sub>Sn<sub>2</sub> films at a series of magnetic fields. A representative sequence during the magnetization reversal is shown in Fig. 4(d). Starting at -10.0 mT, the magnetization is in a saturated state along -X (blue). The reversal initiates with the nucleation of white regions with  $M_x \approx 0$ , mainly at the edges of the sample. This can be explained by the minimization of domain wall energy as the edge boundary does not contribute a domain wall energy cost. The nucleation at the edges initiates magnetization reversal, which results in a more rounded hysteresis loop compared to the uniform films. With increasing magnetic field, the domains of opposite polarity grow inward and coalesce across the channel. At about +1.5 mT, the magnetic structure is in a multidomain state with characteristic features (e.g., blue and red regions) ranging from 1 to 10  $\mu$ m in size. By +2.5 mT, most of the magnetic moments have switched to +Xdirection, with only a few regions remaining along -X. Finally, at +10.0 mT, the magnetization reversal is complete and the films are fully saturated along +X.

Finally, to demonstrate the ability to control the sample structure at the atomic level, we synthesized a  $[Fe_3Sn_2(2 \text{ nm})/Fe_3Sn (2 \text{ nm})]_5$  superlattice using the AL-MBE technique. The  $[Fe_3Sn_2/Fe_3Sn]_5$  superlattice samples were grown under the same conditions as  $Fe_3Sn_2$  samples but with different atomic layer deposition sequences. For a 2 nm Fe\_3Sn layer, we deposit nine



**FIG. 4.** Anomalous Nernst effect imaging. (a) Schematics of thermal gradient microscopy. The laser beam is scanned over the sample surface, and the induced local ANE voltage reflects the local magnetic properties. (b) Microscope image of a typical device [the dashed rectangle corresponds to the area imaged in (d)]. (c) ANE hysteresis loop of a 20 nm Fe<sub>3</sub>Sn<sub>2</sub> film. (d) Magnetization reversal of a 20 nm film through multidomain state imaged by ANE at a series of magnetic fields  $\mu_0 H_x$ .



**FIG. 5.**  $[Fe_3Sn_2/Fe_3Sn]_5$  superlattice. (a) RHEED pattern of  $[Fe_3Sn_2/Fe_3Sn]_5$  superlattice. (b) TEM image of  $[Fe_3Sn_2/Fe_3Sn]_5$  superlattice. (c) EDX of  $[Fe_3Sn_2/Fe_3Sn]_5$  superlattice.

atomic layers of Fe<sub>3</sub>Sn without any Sn<sub>2</sub> spacers. For a 2 nm Fe<sub>3</sub>Sn<sub>2</sub> layer, we deposit two atomic layers of Fe<sub>3</sub>Sn and one atomic layer of Sn<sub>2</sub> and repeat a total of three times. The RHEED pattern of a [Fe<sub>3</sub>Sn<sub>2</sub>/Fe<sub>3</sub>Sn]<sub>5</sub> superlattice along the [1120] direction of c-sapphire is shown in Fig. 5(a). The XRD data of the [Fe<sub>3</sub>Sn<sub>2</sub>/Fe<sub>3</sub>Sn]<sub>5</sub> superlattice are shown in the supplementary material, Sec. 2.

Such control of the stacking sequence with atomic level precision is confirmed by the HAADF STEM image of the  $[Fe_3Sn_2/Fe_3Sn]$ superlattice structure in Fig. 5(b). From the STEM image, an alternating sequence of 2 nm Fe\_3Sn\_2 (false colored in green) and 2 nm Fe\_3Sn (false colored in red) can be observed with atomic resolution. Within 2 nm Fe\_3Sn\_2 layer, a repetitive stacking of two Fe\_3Sn atomic layers and one Sn\_2 atomic layer can be observed. In contrast, we can only see Fe\_3Sn atomic layers in 2 nm Fe\_3Sn layer. The STEM-EDX chemical map further reveals the repetition of the  $[Fe_3Sn_2/Fe_3Sn]_5$  superlattice structure along the growth direction as shown in Fig. 5(c), where a 2 nm Fe\_3Sn\_2 layer has a stronger signal for the Sn element compared to a 2 nm Fe\_3Sn layer.

We studied the magnetic properties of [Fe<sub>3</sub>Sn<sub>2</sub>/Fe<sub>3</sub>Sn]<sub>5</sub> superlattice samples and compared them to 20 nm Fe<sub>3</sub>Sn<sub>2</sub> samples (see the supplementary material, Sec. 6). The [Fe<sub>3</sub>Sn<sub>2</sub>/Fe<sub>3</sub>Sn]<sub>5</sub> superlattice samples also have easy-plane anisotropy with square hysteresis loops, and very weak anisotropy within the sample plane, as shown by MOKE measurements. However, the longitudinal MOKE signal amplitude for the [Fe<sub>3</sub>Sn<sub>2</sub>/Fe<sub>3</sub>Sn]<sub>5</sub> superlattice is slightly larger than 20 nm Fe<sub>3</sub>Sn<sub>2</sub>. The SQUID measurements show that the [Fe<sub>3</sub>Sn<sub>2</sub>/Fe<sub>3</sub>Sn]<sub>5</sub> superlattice samples have larger out-of-plane saturation field (~1.7 T) and larger saturation magnetization (2.2  $\mu_B$ /Fe). This can be understood because Fe<sub>3</sub>Sn has larger saturation magnetization and larger out-of-plane saturation field than Fe<sub>3</sub>Sn<sub>2</sub>.

In conclusion, we report the atomic layer epitaxy growth of kagome ferromagnet  $Fe_3Sn_2$  thin films on  $Pt(111)/Al_2O_3(0001)$  at low temperatures. The high quality of epitaxial  $Fe_3Sn_2$  films is confirmed by *in situ* RHEED, XRD, AFM, and TEM. Low temperature growth helps to generate a sharp interface between  $Fe_3Sn_2$  and Pt layers, which has been observed by EDX. The magnetic properties

are investigated by magneto-optical Kerr effect, SQUID magnetometry, and anomalous Nernst effect, confirming the easy-plane magnetic anisotropy of the thin films. Using ANE microscopy, we successfully resolve the local in-plane oriented micrometer size domains during magnetization reversal. Finally, we demonstrate the ability to control the sample structure at the atomic level by synthesizing  $[Fe_3Sn_2/Fe_3Sn]_5$  superlattices and confirming their structure by TEM. These advances enable novel heterostructures for exploring the rich physics of kagome magnets.

See the supplementary material for additional AFM, XRD, EELS, MOKE, and SQUID data.

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#### AUTHOR DECLARATIONS

#### **Conflict of Interest**

The authors have no conflicts to disclose.

#### **Author Contributions**

Shuyu Cheng: Conceptualization (lead); Investigation (lead); Writing – original draft (lead). Binbin Wang: Investigation (lead); Writing – original draft (supporting). Igor Lyalin: Conceptualization (lead); Investigation (lead); Writing – original draft (lead). Núria Bagués: Investigation (supporting); Writing – original draft (supporting). Alexander J. Bishop: Investigation (supporting); Writing – original draft. David W. McComb: Supervision (lead); Writing – original draft (lead). **Roland K. Kawakami:** Conceptualization (lead); Supervision (lead); Writing – original draft (lead).

#### DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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