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Spin excitations in the Kagome-lattice metallic antiferromagnet $\text{Fe}_{0.89}\text{Co}_{0.11}\text{Sn}$

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Kagome-lattice materials have attracted tremendous interest due to the broad prospect for seeking superconductivity, quantum spin liquid states, and topological electronic structures. Among them, the transition-metal Kagome lattices are high-profile objects for the combination of topological properties, rich magnetism, and multiple orbital physics. Here we report an inelastic neutron scattering study on the spin dynamics of a Kagome-lattice antiferromagnetic metal $\text{Fe}_{0.89}\text{Co}_{0.11}\text{Sn}$. Although the magnetic excitations can be observed up to ~ 250 meV, well-defined spin waves are only identified below ~ 90 meV, and can be modeled using Heisenberg exchange with ferromagnetic in-plane nearest-neighbor coupling (J_1), in-plane next-nearest-neighbor coupling (J_2), and antiferromagnetic (AFM) interlayer coupling (J_c) under linear spin-wave theory. Above ~ 90 meV, the spin waves enter the itinerant Stoner continuum and become highly damped particle-hole excitations. At the K point of the Brillouin zone, we reveal a possible band crossing of the spin wave, which indicates a potential Dirac magnon. Our results uncover the evolution of the spin excitations from the planar AFM state to the axial AFM state in $\text{Fe}_{0.89}\text{Co}_{0.11}\text{Sn}$, solve the magnetic Hamiltonian for both states, and confirm the significant influence of the itinerant magnetism on the spin excitations.

I. INTRODUCTION

The magnetic Kagome lattice, a two-dimensional (2D) network of corner-sharing triangles surrounding hexagons, provides an ideal platform to search for exotic states such as quantum spin liquids [1–4] and other topological quantum states [5–14]. Theoretical studies have shown that the typical electronic bands of Kagome lattices contain linearly dispersive Dirac bands and nondispersive flat bands [15, 16]. When a Kagome lattice is occupied by $3d$ transition-metal atoms, the combination of the rich magnetism, topological electronic bands, and multiple orbital characteristics will induce abundant novel phenomena such as the anomalous Hall effect [5–9, 11, 12].

In modern magnetic theory, the interaction between electron spins can be described by a local moment picture or itinerant electron model [17–20]. Although the former case is usually appropriate in the magnetic insulators and the latter model always comes into play in metallic magnets, some metallic systems can be analyzed by the local moment model [21, 22]. What's more, both the local moment and the itinerant electron scenarios can coexist in some systems [23–25]. For example, in iron-based superconductors, the spin waves can be reproduced by an effective Heisenberg Hamiltonian with considering the anisotropic spin-wave damping characteristics of an itinerant electron system [23, 24]. In some itinerant magnets,

well-defined spin-wave excitations only can be observed in the low-energy/long-wavelength region before entering the Stoner continuum [Fig. 1(e)], in which the spin waves decay into damped particle-hole excitations. [18–21, 26–28].

Recently, Dirac fermions and flat electronic bands have been reported in Kagome-lattice metallic antiferromagnet FeSn, paramagnet CoSn, and the doped compounds $\text{Fe}_{1-x}\text{Co}_x\text{Sn}$ by angle-resolved photoemission spectroscopy studies [10, 13, 29, 30]. The FeSn/CoSn family has a hexagonal structure with $P6/mmm$ space group. The Fe/Co atoms form the Kagome lattice with hexagonal holes filled with Sn atoms [Fig. 1(a)]. In antiferromagnetic (AFM) FeSn, below the $T_N = 365$ K, the magnetic moments of Fe in each Kagome layer align ferromagnetically, and the adjacent ferromagnetic (FM) layers stack antiparallely along the c axis [Fig. 1(b)] [13, 29, 31–34]. With Co substitution at the Fe site in FeSn, the ordered moments' direction can be tuned from that in the ab plane (planar AFM) [Fig. 1(b)] to along the c axis (axial AFM) [Fig. 1(c)] continuously by crossing an intermediate state (tilted AFM) [35, 36]. During this process, the magnetic moments of the neighboring FM layers remain antiparallel to each other. At some specific levels of Co doping, these different AFM states can be obtained by simply changing the temperature [35].

Although the recent inelastic neutron scattering (INS) studies on FeSn show some differences, both reports confirm the non-negligible effect of itinerant electrons on the spin excitations [37, 38], which suggests that the combination of localized and itinerant magnetism should be considered in this kind of metallic Kagome-lattice AFMs. While theoretical calculation suggested the existence of a

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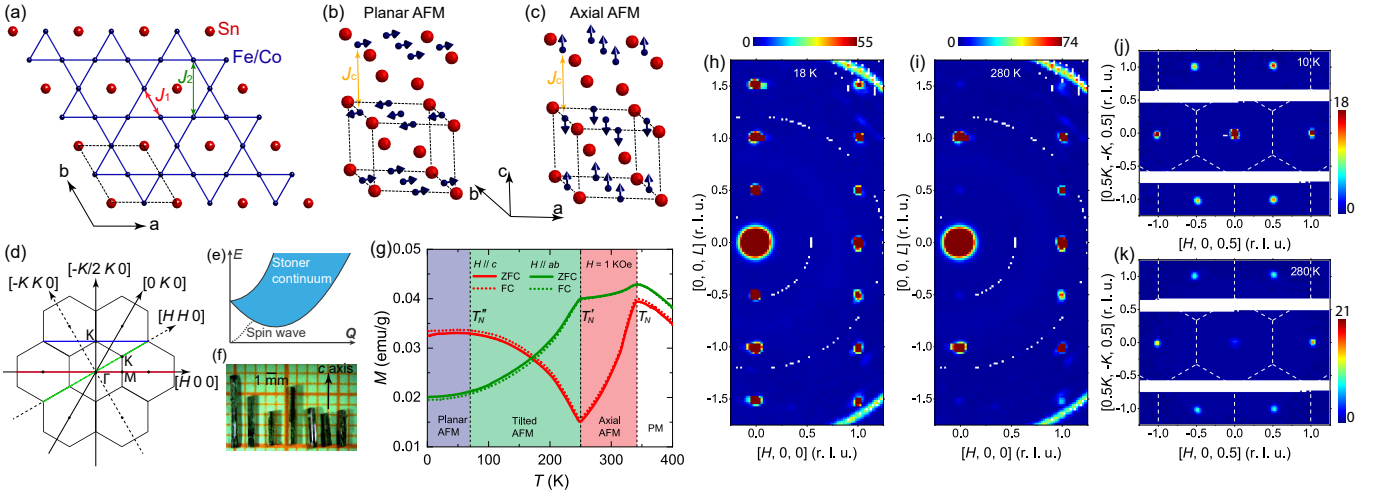


FIG. 1. (a) The Fe/Co-Kagome layer with the hexagonal holes filled with Sn in $\text{Fe}_{1-x}\text{Co}_x\text{Sn}$. The in-plane nearest-neighbor and the next-nearest-neighbor exchange couplings are indicated by the red and green arrows, respectively. (b)-(c) The magnetic structures for the planar and axial AFM in $\text{Fe}_{1-x}\text{Co}_x\text{Sn}$. The orange arrows represent the interlayer nearest-neighbor exchange coupling J_c . (d) The schematic of the 2D Brillouin zone of $\text{Fe}_{1-x}\text{Co}_x\text{Sn}$. (e) The schematic of the spin wave and Stoner continuum in some metallic magnets. (f) A typical photo of $\text{Fe}_{0.89}\text{Co}_{0.11}\text{Sn}$ single crystals on 1-mm grid paper. The long axis of rod-like crystals is the crystallographic c axis. (g) The magnetization as a function of temperature of our $\text{Fe}_{0.89}\text{Co}_{0.11}\text{Sn}$ sample. (h)-(i) Zero-energy ($E = 0 \pm 0.2$ meV) 2D slices in $(H 0 L)$ with $E_i = 10$ meV at 18 K (planar AFM state) and 280 K (axial AFM state), respectively. The arcs at the lower right corner and the upper right corner are scattering intensity from the aluminum sampler holder. (j)-(k) Zero-energy ($E = 0 \pm 1$ meV) 2D slices in $(H K 0.5)$ with $E_i = 80$ meV at 10 K (planar AFM state) and 280 K (axial AFM state), respectively. The dashed lines indicate the boundary of the Brillouin zones in the $(H K 0)$ plane.

69 magnetic flat band in FeSn [37], INS studies did not observe it [37, 38]. In addition, a damped Dirac magnon has
 70 been suggested to exist in FeSn [38]. The Co substitution
 71 in FeSn enriches the magnetism and may change the itinerancy of the electrons [34, 35], which makes $\text{Fe}_{1-x}\text{Co}_x\text{Sn}$
 72 a good candidate to study the topological magnon, magnetic flat band, and their interplay with the itinerant
 73 electrons as well as the evolution of these properties with
 74 spin orientations.

78 In this study, we select $\text{Fe}_{0.89}\text{Co}_{0.11}\text{Sn}$, which contains
 79 axial AFM, tilted AFM, and planar AFM states in different temperature regions, as our research object. By
 80 employing magnetization and neutron scattering measurements, we first confirm the existence of the different AFM
 81 states in $\text{Fe}_{0.89}\text{Co}_{0.11}\text{Sn}$. We subsequently obtain the in-plane FM spin excitations from zero energy to ~ 250 meV
 82 together with the out-of-plane AFM spin wave below ~ 25
 83 meV in both planar and axial AFM states, which suggests
 84 quasi-2D magnetism in $\text{Fe}_{0.89}\text{Co}_{0.11}\text{Sn}$. The magnetic excitations below ~ 80 – 90 meV can be described by linear
 85 spin-wave theory (LSWT) simulation. Above ~ 90 meV,
 86 the spin waves enter the Stoner continuum and decay into
 87 the highly damped particle-hole excitations. Evidence
 88 of the existence of the Dirac magnon is also observed at
 89 the K point of the Brillouin zone (BZ), albeit its upper
 90 part is obscured due to the interaction with the Stoner
 91 continuum from itinerant magnetism.

66 II. EXPERIMENTAL DETAILS

97 We prepared high-quality single crystals of
 98 $\text{Fe}_{0.89}\text{Co}_{0.11}\text{Sn}$ using the self-flux method. Details
 99 can be found in supplementary information (SI) [39]
 100 (see, also, references [40, 41] therein). The crystals are
 101 long bars along the crystalline c axis with a hexagonal
 102 cross-section [Fig. 1(f)]. Magnetization measurements
 103 were performed using a Quantum Design (QD) Mag-
 104 netic Properties Measurement System (MPMS3). We
 105 co-aligned about 100 single crystals in the $(H 0 L)$
 106 scattering plane on thin aluminum plates to obtain a
 107 mosaic sample with a mass of about 2 g and mosaicity
 108 below 1.5° [39]. The neutron scattering experiments were
 109 performed on the time-of-flight Wide Angular-Range
 110 Chopper Spectrometer (ARCS) [42], Fine-Resolution
 111 Fermi Chopper Spectrometer (SEQUOIA) [43], and
 112 Cold Neutron Chopper Spectrometer (CNCS) [44, 45]
 113 at the Spallation Neutron Source, Oak Ridge National
 114 Laboratory. Measurements were carried out with a series
 115 of incident neutron energies $E_i = 3.32$ meV, 10 meV, 80
 116 meV, 150 meV, 250 meV, 300 meV, and 400 meV in both
 117 the planar AFM (at $T = 10$ K and 18 K) and axial AFM
 118 (at $T = 280$ K) states. The sample was rotated along the
 119 vertical axis in a wide angle range (except for the $E_i =$
 120 400 meV, where the beam was fixed with $k_i \parallel c$ axis) to
 121 make a complete survey in the energy and momentum
 122 space. We used the software packages MANTID [46]
 123 and HORACE [47] for neutron scattering data reduction

and analysis. The neutron scattering intensities are normalized to a same scale with arbitrary units using the incoherent elastic scatterings of the sample [39]. In the whole paper, a wave vector \mathbf{Q} will be shown in reciprocal lattice units (r. l. u.), in which $\mathbf{Q} = (H, K, L)$ means $\mathbf{Q} = H\mathbf{a}^* + K\mathbf{b}^* + L\mathbf{c}^*$, where \mathbf{a}^* , \mathbf{b}^* , and \mathbf{c}^* are basis vectors in reciprocal space.

III. NEUTRON SCATTERING RESULTS

A. Elastic neutron scattering

The magnetization measurements of $\text{Fe}_{0.89}\text{Co}_{0.11}\text{Sn}$ show three characteristic temperatures, named $T_N \approx 340$ K, $T'_N \approx 250$ K, and $T''_N \approx 70$ K [Fig. 1(g)], which correspond to the phase-transition temperatures from paramagnetic (PM) state to axial AFM state, axial AFM state to tilted AFM state, and tilted AFM to planar AFM state, respectively [35]. In order to confirm the magnetic phases in our sample, we first check the elastic neutron scattering results and then compare the results with that in Ref. [35].

Figures 1(h)–1(k) present several zero-energy 2D slices in $(H\ 0\ L)$ and the $(H\ K\ 0.5)$ planes. In Fig. 1(h), we can see strong peaks appear at $Q = (0, 0, n/2)$ ($n =$ integer) and $Q = (1, 0, n/2)$. The peaks at integer L are nuclear peaks, while the peaks at half-integer L are magnetic Bragg peaks, which correspond to a propagation vector $\mathbf{q} = (0, 0, 1/2)$. The peaks in $(H\ K\ 0.5)$ plane shown in Fig. 1(j) further confirm the fact that the ordered moments align ferromagnetically in the ab plane. These observations are consistent with the previous neutron diffraction results [35]. Since neutron scattering measurements probe the magnetic moment components that are normal to the wave vector \mathbf{Q} , we expect no magnetic Bragg peaks at $Q = (0, 0, n + 1/2)$ for an axial AFM state due to the parallel direction between magnetic moments and \mathbf{Q} s [Fig. 1(c)]. However, weak magnetic Bragg peaks still can be observed at $Q = (0, 0, n + 1/2)$ at $T = 280$ K [Figs. 1(i) and 1(k)]. A similar phenomenon has been reported in Ref. [35], and was explained as the tails of inelastic scattering by low-energy transverse magnons. In our study, we can rule out this possibility in $\text{Fe}_{0.89}\text{Co}_{0.11}\text{Sn}$ clearly (see details in subsection B), and confirm the existence of the small in-plane magnetic moment components which result in the weak magnetic peaks at $Q = (0, 0, n + 1/2)$ in the axial AFM state. At last, we calculate the component of the ordered moment along c axis $m_c \approx 1.39 \mu_B$ and the small in-plane component $m_{ab} \approx 0.12 \mu_B$, which corresponds to a small canting angle ($\sim 4.84^\circ$) of the ordered spins away from the c axis (see details in SI [39]).

B. Low- and intermediate-energy spin wave

Now the spin dynamics of $\text{Fe}_{0.89}\text{Co}_{0.11}\text{Sn}$ are discussed. Figure 2 presents the spin wave results in the low- and intermediate-energy range, which were collected with $E_i = 3.32$ meV (at CNCS), $E_i = 80$ meV (at SEQUOIA), and $E_i = 150$ meV (at ARCS, only $T = 10$ K). There are three magnetic (Fe/Co) atoms in one unit cell [Fig. 1(b) and 1(c)], which will give rise to three magnon branches. As shown in Figs. 2(a) and 2(d), we can see the steep dispersion of the acoustic magnon along $[H, 0, 0.5]$ direction for both planar (10 K) and axial (280 K) AFM states. While the top of the acoustic spin wave band of the planar AFM state cannot be clearly seen in the measured energy range with $E_i = 80$ meV, the energy band top for the axial AFM state seems to appear at ~ 67 meV. Figure 2(g) shows spin wave dispersion along $[H, 0] \rightarrow [H, H]$ path at 10 K measured with $E_i = 150$ meV. The top of the acoustic spin wave band at the M point appears around 82 meV, above which the weak spin excitation intensity continues up to 130 meV. On the other hand, the spin wave dispersion along the $[0, 0, L]$ direction reaches the band top at about 21 meV and 13 meV for the planar and axial AFM state, respectively [Figs. 2(b) and 2(e)]. We extracted 1D constant-energy curves from the spin-wave dispersion in Figs. 2(a)–2(b), 2(d)–2(e), and fitted the curves with Gaussian functions. Some of the 1D constant-energy curves are shown in Figs. 2(h)–2(i). The data points in Figs. 2(a), 2(d), and 2(g) are the peak positions of these 1D constant-energy curves.

In order to figure out whether the spin wave of $\text{Fe}_{0.89}\text{Co}_{0.11}\text{Sn}$ is gapped or gapless, we measured the low energy excitations with $E_i = 3.32$ meV (with an energy resolution ~ 0.11 meV at the elastic position [39]). From Figs. 2(c) and 2(f), we can see sharp spin waves stem from $Q = (0, 0, 0.5)$ for both states. The 1D energy cuts at $Q = (0, 0, 0.5)$ show the evolution of the spin wave intensity more intuitively [Fig. 2(j)]. At 10 K, we do not see the abrupt intensity decrease with the decreasing energy at $Q = (0, 0, 0.5)$, which was observed and considered as evidence of the spin gap in FeSn [37]. This indicates the spin wave of the planar AFM state in $\text{Fe}_{0.89}\text{Co}_{0.11}\text{Sn}$ is gapless in our resolution limit. At 280 K, the intensity of the spin wave first gradually increases with the decreasing energy above 1 meV, then abruptly decreases with the decreasing energy below ~ 0.5 meV until the spin wave touches the tail of the Bragg peak, and upturns below ~ 0.1 meV [Fig. 2(j)]. Having this high-resolution data at the axial AFM state, two important issues can be figured out clearly. First, in the axial AFM state, there is a small spin gap below ~ 0.5 meV, although the gap is not fully opened. Second, the weak magnetic Bragg peaks observed at $Q = (0, 0, n + 1/2)$ (see subsection A) are intrinsic magnetic peaks for the existence of in-plane magnetic moments in the axial AFM state. These peaks cannot be the tails of inelastic scattering by low-energy transverse magnons [35]. Because the inelastic tail should have a rather low intensity below ~ 0.2 meV for the opening of

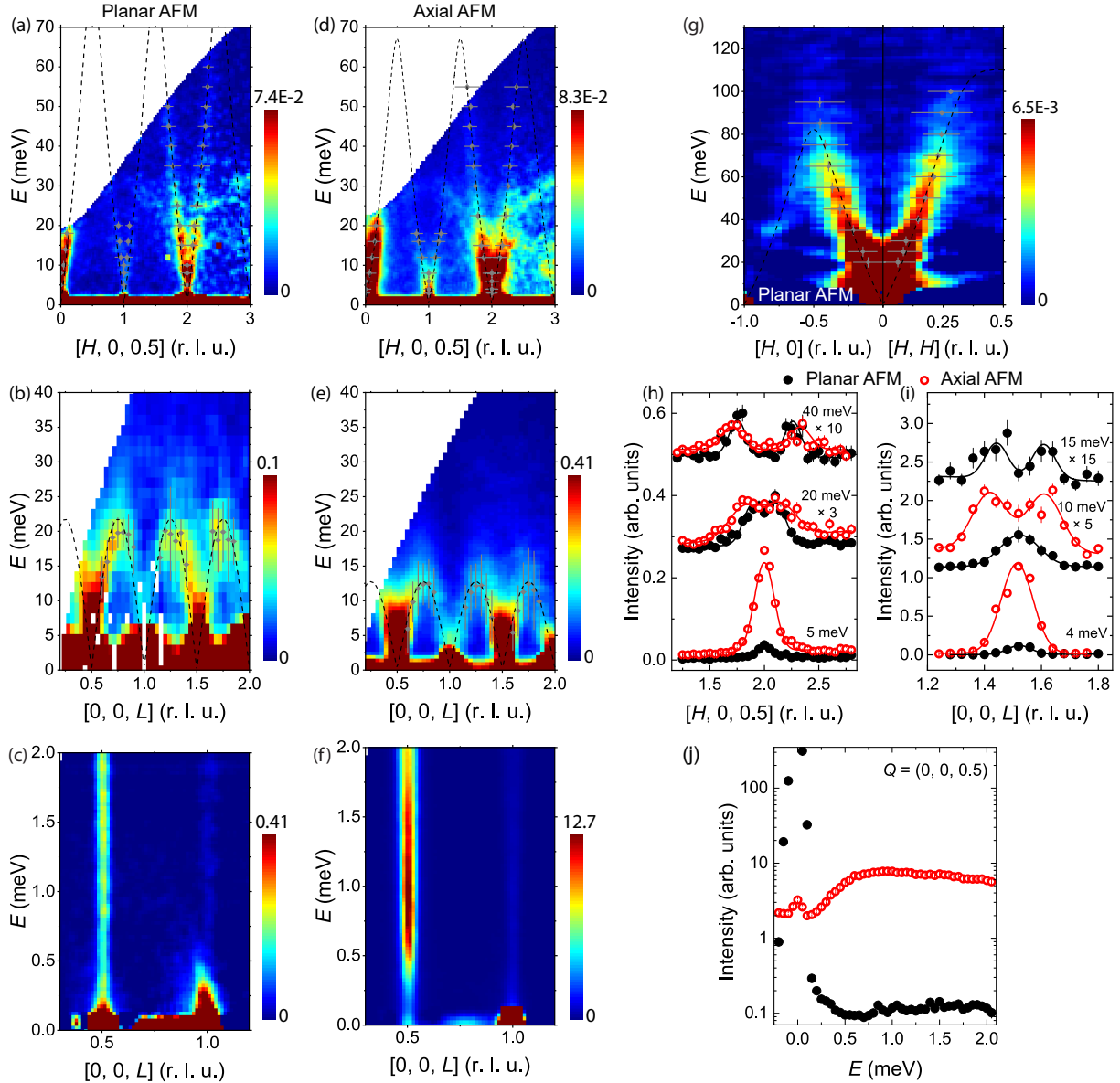


FIG. 2. Dispersion of spin wave. (a)–(b) Spin-wave dispersion along the $[H, 0, 0.5]$ [the red path in Fig. 1(d)] measured with $E_i = 80$ meV, and along the $[0, 0, L]$ direction measured with $E_i = 150$ meV, $T = 10$ K. (c) Spin-wave dispersion along the $[0, 0, L]$ direction measured with $E_i = 3.32$ meV, $T = 10$ K. (d)–(e) Spin-wave dispersion along the $[H, 0, 0.5]$ and $[0, 0, L]$ measured with $E_i = 80$ meV, $T = 280$ K. (f) Spin-wave dispersion along the $[0, 0, L]$ direction measured with $E_i = 3.32$ meV, $T = 280$ K. The extra intensities in (a) and (d) around 10 meV and 30 meV at high Q region are phonon signal. (g) Spin-wave dispersion along $[H, 0] \rightarrow [H, H]$ path at 10 K measured with $E_i = 150$ meV. The extra intensity around 10 meV is the intensity of phonon scattering. (h)–(i) 1D constant-energy curves along the $[H, 0, 0.5]$ direction and $[0, 0, L]$ direction at different energies in both planar and axial AFM states. The solid lines are fittings with the Gauss function. (j) 1D constant-momentum cuts at $Q = (0, 0, 0.5)$ in both planar and axial AFM states. The integration range along $[-K/2, K/2]$ direction for all the panels is $K = [-0.1, 0.1]$. The broken lines in panels (a), (b), (d), (e), and (g) are results of LSWT fittings (see details in next section). The data points with error bars in panels (a), (d) and (g) are the peak positions of the 1D constant-energy curves as that shown in (h)–(i). The vertical error bars are the energy resolution of the instrument, and the horizontal error bars are the full width at half maximum (FWHM) of the Gauss fittings there. The data points in panels (b) and (e) are peak positions of the 1D constant-momentum curves. The vertical error bars are the line width (FWHM) of spectra, and the horizontal error bars are integration momentum range.

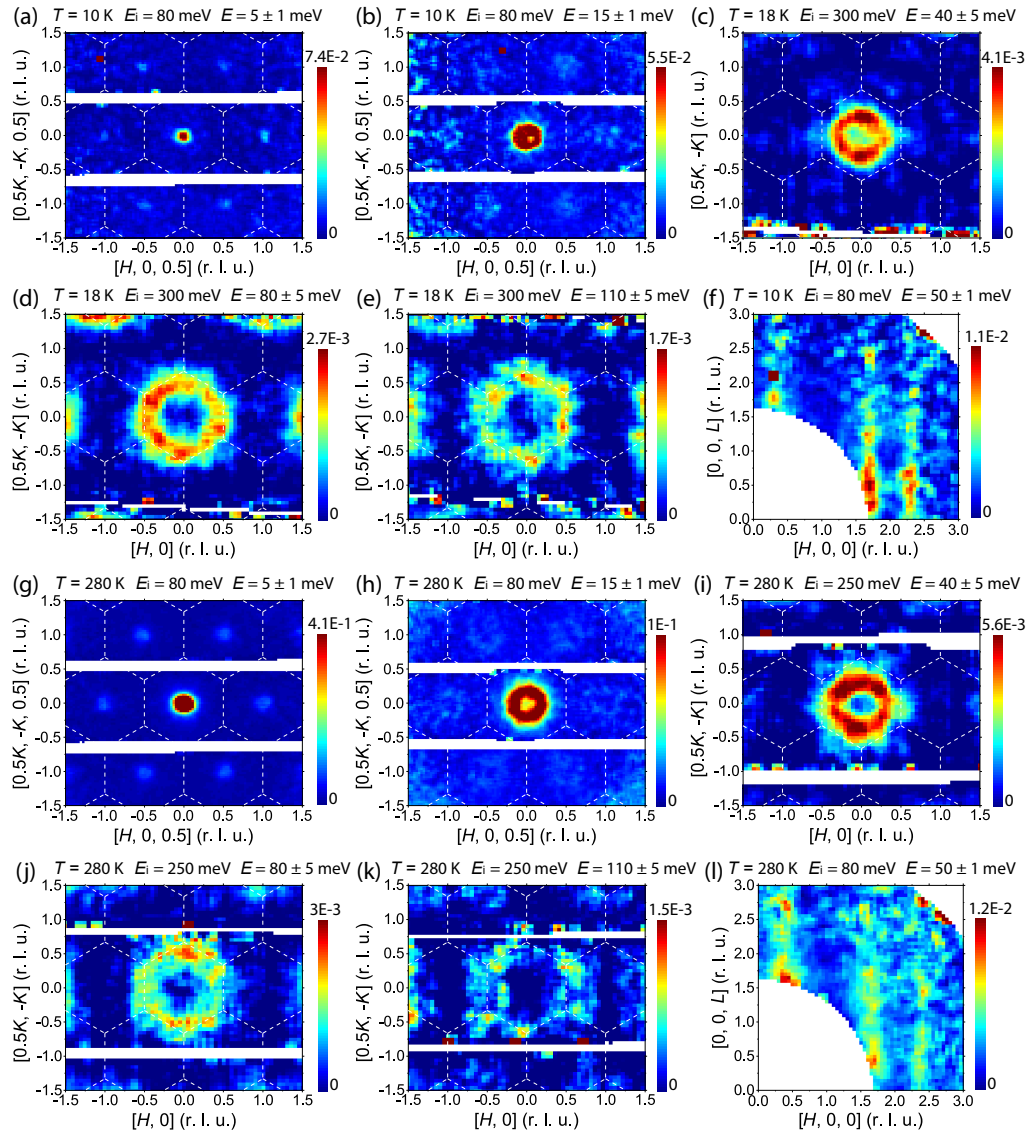


FIG. 3. Constant-energy 2D slices in the $(H K)$ plane and $(H 0 L)$ plane. (a)–(e) The evolution of the spin wave with the increasing energy in the $(H K)$ plane at $T = 10$ K (planar AFM state). (f) Constant-energy slice in $(H 0 L)$ plane at $T = 10$ K and $E = 50 \pm 1$ meV. (g)–(k) The evolution of the spin wave with the increasing energy in the $(H K)$ plane at $T = 280$ K (axial AFM state). (l) Constant-energy slice in $(H 0 L)$ plane at $T = 280$ K and $E = 50 \pm 1$ meV. The white broken lines represent the boundary of Brillouin zones.

230 the spin gap. If the elastic peaks at $Q = (0, 0, n + 1/2)$ 239
 231 are inelastic tails, their intensity should be lower than (or
 232 comparable with) the intensity of the excitations at ~ 0.2
 233 meV. With this, there cannot be the obviously upward
 234 intensity below ~ 0.1 meV as shown in Fig. 2(j). Thus,
 235 we demonstrate that the weak magnetic Bragg peaks ob-
 236 served at $Q = (0, 0, n + 1/2)$ at 280 K are intrinsic, and
 237 the magnetic moments are not perfectly aligned along the
 238 c axis in the axial AFM state of $\text{Fe}_{0.89}\text{Co}_{0.11}\text{Sn}$.

C. High-energy spin excitations

240 To cover the high-energy spin excitations of
 241 $\text{Fe}_{0.89}\text{Co}_{0.11}\text{Sn}$, we measured the spin dynamics with
 242 higher incident neutron energies: $E_i = 300$ meV (at
 243 ARCS and $T = 18$ K) and 250 meV (at SEQUOIA and
 244 $T = 280$ K). Figure 3 presents some constant-energy 2D
 245 slices in the $(H K)$ plane and $(H 0 L)$ plane. In the $(H K$
 246 $0.5)$ plane, the spin waves stem from the same positions
 247 where the magnetic Bragg peaks are observed in Figs. 1(j)
 248 and 1(k). The small spots then evolve to be circles with
 249 increasing energy. From Figs. 3(a)–(c) and (g)–(i), we can
 250 see the sizes of the spots/circles in the axial AFM state

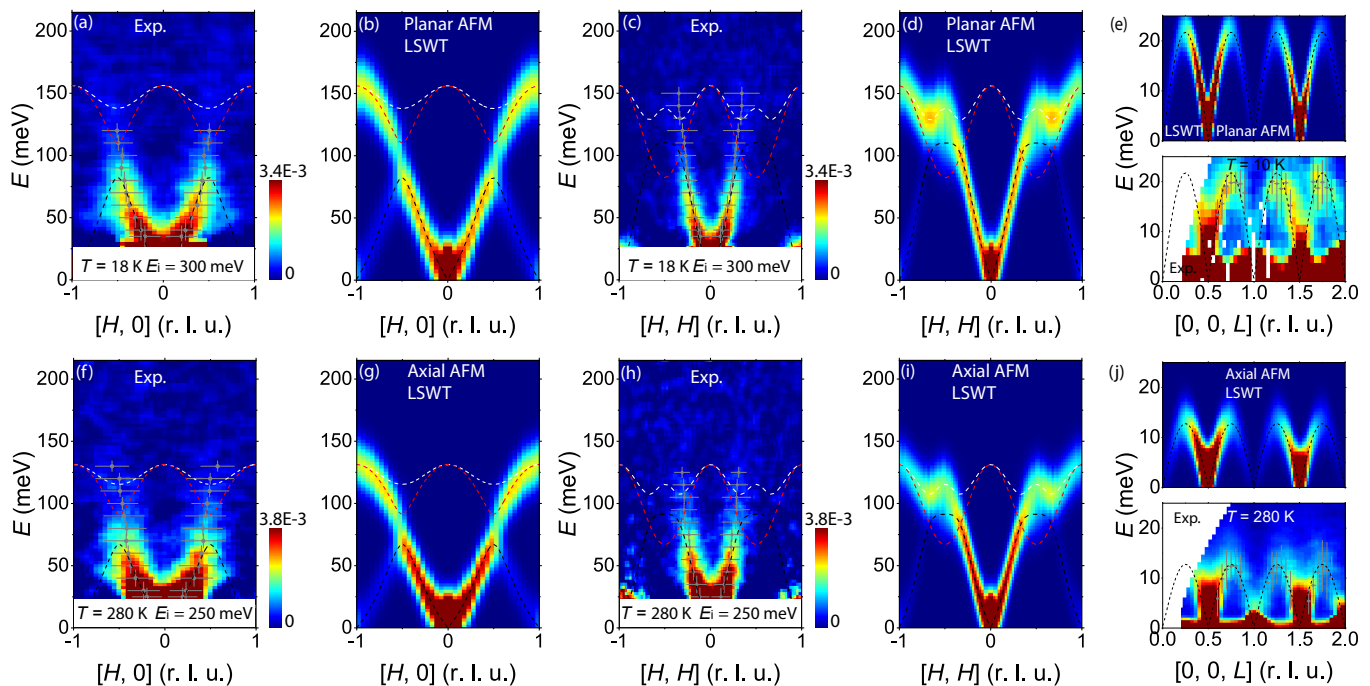


FIG. 4. The high-energy spin wave and LSWT fittings with SpinW. (a)–(d) Experimental INS spectra along the $[H, 0]$ [the red path in Fig. 1(d)] and $[H, H]$ [the green path in Fig. 1(d)] directions measured with $E_i = 300$ meV, $T = 18$ K and the corresponding LSWT calculations. (e) The comparison of the experimental and LSWT calculation of the spin-wave dispersion along $[0, 0, L]$ for the planar AFM state. (f)–(i) INS spectra along the $[H, 0]$ and $[H, H]$ directions measured with $E_i = 250$ meV, $T = 280$ K and the corresponding LSWT calculations. (j) The comparison of the experimental and LSWT calculation of the spin-wave dispersion along $[0, 0, L]$ for the axial AFM state. Some extra intensities that are away from the main dispersion around 50 meV in panels (a), (c), (f), and (h) are residual intensities due to the imperfect background subtraction. The gray data points with error bars in panels (a), (c), (f), and (h) are fitted peak positions of constant-energy curves. The vertical error bars are the energy resolution of the instrument, and the horizontal error bars are the FWHM of the Gauss fittings. The dashed lines are LSWT calculations with the best fitting parameters in Table I, the black lines indicate the acoustic magnons, and the red and white dashed lines indicate the optical magnons.

(280 K) are always larger than that in the planar AFM state (10 K or 18 K), which means the spin excitation can reach the BZ boundary at a lower energy in the axial AFM state, and is consistent with the analysis in subsection B. When the neutron energy transfer approaches ~ 80 meV, the spin excitations evolve to the edge of the BZs, which indicates the energy top of the acoustic magnon band. Furthermore, we found that the spin excitation has no obvious intensity modulation along the $[0, 0, L]$ direction for energies above ~ 30 meV [Fig. 3(f) and 3(l)]. Thus, the results above 30 meV shown in Figs. 3 and 4 were extracted by integrating a wide range of L ($-5 \leq L \leq 5$), and we will omit the L indices for these cases.

The INS spectra up to 215 meV are shown in Fig. 4. For the planar AFM state, the dispersion along the $[H, 0]$ direction shows strong intensity below ~ 100 meV, above which the signals become diffusive and rather weak, but still can be identified up to ~ 200 meV [Fig. 4(a)]. For the dispersion along $[H, H]$ direction [Fig. 4(c)], a sharp spin wave below ~ 100 meV and an obvious intensity decrease above ~ 100 meV are also observed. In the axial AFM state [Fig. 4(f) and 4(h)], the high-energy spin excitations are similar to those in the planar AFM

state, while the energy scale is smaller than in the planar AFM state. It is worth noting that here we only can observe the clear acoustic spin wave mode and the weak diffusive spin excitations (between ~ 90 and 200 meV), any indications of the other two expected magnon modes cannot be identified.

IV. LSWT SIMULATIONS AND DISCUSSIONS

To understand the experimentally observed magnetic excitations, we employ the LSWT simulations using SpinW library [48]. We use the following Heisenberg Hamiltonian:

$$\mathcal{H} = J_1 \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{\langle\langle i,j \rangle\rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_c \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + K_c \sum_{\langle i \rangle} (S_i^z)^2 + K_a \sum_{\langle i \rangle} (S_i^{\hat{e}_x})^2, \quad (1)$$

where J_1 is the in-plane nearest-neighbour (NN) exchange coupling, J_2 is the in-plane next-nearest-neighbour (NNN) exchange coupling, J_c is the NN interlayer exchange

coupling. The last two terms represent the single-ion anisotropy. It should be noted that the last term is only applicable for the axial AFM state. With $K_a < 0$, $K_a(S_i^{\hat{r}})^2$ represents the in-plane easy-axis anisotropy following the lattice symmetry, which is responsible for the small canting angle of the ordered spins. The in-plane easy-axis direction \hat{e}_r depends on the position (\mathbf{r}) of the magnetic atoms (see details in SI [39]). Due to the itinerant properties of such a metallic system, the spin value could be ambiguous. The effective spin value may also change from the planar AFM to axial AFM state. To describe the magnetic Hamiltonian smoothly, we thus use the combination of the spin value and exchange coupling parameters SJ_1 , SJ_2 , SJ_c , SK_c , and SK_a hereinafter.

We first cut the experimental spectra and fitted them to get the dispersion relation and intensity of the spin wave. Then we fit the extracted data using SpinW with Hamiltonian (1) to get the exchange coupling parameters (see details in SI [39]). The best fitting parameters are summarized in Table I. The fitted spin wave dispersion curves for the acoustic magnon are shown as dashed lines in Figs. 2(a)–2(b), 2(d)–2(e), and 2(g), which indeed can describe the data points from the experimental results perfectly. The calculated dynamical spin structure factors with the parameters in Table I are shown in Figs. 4(b), 4(d), 4(e), 4(g), 4(i) and 4(j).

Note that the single-ion anisotropy parameters SK_c and SK_a in Table I are determined separately from SJ_1 , SJ_2 and SJ_c [39]. This is a reasonable approach since the anisotropy terms are rather small and have negligible influence on the spin wave dispersion here. Although no spin gap can be identified in the planar AFM state, it is still necessary to include an easy-plane single-ion anisotropy ($SK_c > 0$) to confine the ordered spins in the ab plane. As for the axial AFM state, a minor spin gap below 0.5 meV has been identified, which requires a non-zero single-ion anisotropy to open the gap. However, as the small canting angle ($\sim 4.84^\circ$) has been confirmed by our previous analysis in section III.A-B, a single easy-axis anisotropy term ($SK_c < 0$) cannot stabilize such a special magnetic structure. Our solution is to add the $K_a \sum_{\langle i \rangle} (S_i^{\hat{r}})^2$ term ($SK_a < 0$) in the Hamiltonian. At last, we estimate the single-ion anisotropy parameters to be $0 < SK_c \leq 0.0038$ meV in the planar AFM state, and $SK_c = SK_a = -0.009$ meV in the axial AFM state (see details in SI [39]).

From Table I, we can see that the generalized exchange coupling parameters SJ_1 , SJ_2 and SJ_c decrease on different levels from the planar state to the axial AFM state. Specifically, $(SJ_1)_{axial}/(SJ_1)_{planar} \approx 0.876$, $(SJ_2)_{axial}/(SJ_2)_{planar} \approx 0.878$, $(SJ_c)_{axial}/(SJ_c)_{planar} \approx 0.586$. If the effective local spin value in the Hamiltonian is supposed to be proportional to the ordered magnetic moment of the ground state, we can get the ratio of the effective spin value between the planar and the axial AFM states: $S^{axial}/S^{planar} = m^{axial}/m^{planar} \approx 0.760$ (see details in SI [39]). Then the ratio of the real exchange coupling parameters: $J_1^{axial}/J_1^{planar} \approx 1.153$,

$J_2^{axial}/J_2^{planar} \approx 1.155$, $J_c^{axial}/J_c^{planar} \approx 0.771$. This means that from the planar to axial AFM state, the in-plane parameters J_1 and J_2 increase synchronously, while the out-of-plane parameter J_c decreases. These interesting evolutions of the effective exchange couplings should reflect the changes of the electronic band structure for the planar-to-axial phase transition, which have been demonstrated by the theoretical calculations of the electronic structure on a similar compound $\text{Fe}_{0.94}\text{Co}_{0.06}\text{Sn}$ [30]. To explain the observed changes of the effective exchange coupling parameters from the change of the electronic structure one should project the exchange coupling interactions into orbital resolved contributions. Exchange coupling parameters can be calculated from the electronic structure using the known formalisms, e.g., local spin density functional [49] or the real-space linear-muffin-tin method [50, 51], which require extensive computational work that is out of the scope of the current work.

By comparing the experimental and the LSWT simulation results, it is clear that the LSWT calculation works well only for the acoustic spin wave. For the spin excitations above the acoustic magnon, the data looks ambiguous, from which we cannot identify the residual two optical magnon modes. The LSWT simulation cannot cover the experimental results. The weak and ambiguous excitations above the acoustic magnon and the disappearance of the optical magnon modes can be explained by the interaction between the spin wave and the Stoner excitations from the itinerant magnetism [18–21, 26–28, 37, 38, 52]. As depicted in Fig. 1(e), we only can observe well-defined spin wave at the relatively low-energy region (below ~ 90 meV here) just before touching the lower boundary of Stoner continuum. After entering the Stoner continuum (above ~ 90 -100 meV here), the spin waves decay into the particle-hole excitations, which makes the optical magnon modes invisible, and only leaves us the observable weak damped excitations up to ~ 250 meV. A recent *ab initio* study on FeSn indicates that the Stoner continuum appears above ~ 80 -100 meV and overlaps with the high-energy magnon spectra, which results in the strong damping of the magnon [52]. Our results in $\text{Fe}_{0.89}\text{Co}_{0.11}\text{Sn}$ here is qualitatively consistent with this calculation and the experimental INS results in FeSn [38].

We further check the data across the K points of BZs [the blue path shown in Fig. 1(d)] in both AFM states. In Fig. 5(a), we can see the spectrum shows a downward cone-like shape with the vertex appearing at ~ 100 meV at $T = 18$ K. Above the downward cone, the excitation intensity becomes weak and diffusive, and is similar to the aforementioned results shown along the red and green paths in Fig. 1(d). Similarly, at $T = 280$ K, a downward cone with a slightly lower vertex (~ 85 meV) can be also identified [Fig. 5(b)], although it is not as clear as that at $T = 18$ K. This feature is not easy to understand at first glance. But if we consider the aforementioned interaction between the spin wave and itinerant Stoner continuum, the downward cone-like excitation here should

TABLE I. Exchange coupling parameters (meV) for the magnetic Hamiltonian (1) obtained from the LSWT fittings

Ordering type	SJ_1	SJ_2	SJ_c	SK_c	SK_a
Planar AFM	-18.15 ± 4.9	-4.50 ± 1.89	10.87 ± 2.21	$0 < K_c \leq 0.0038$	–
Axial AFM	-15.90 ± 4.32	-3.95 ± 1.55	6.37 ± 1.54	-0.009	-0.009

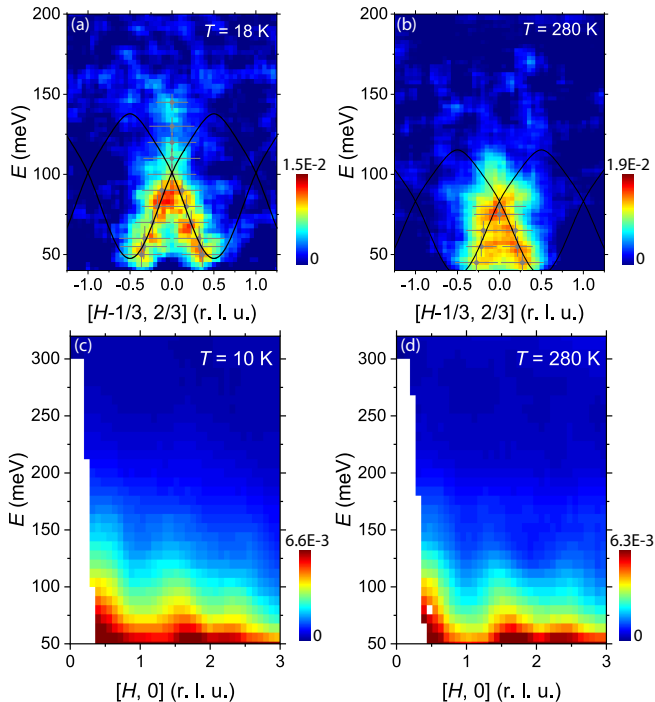


FIG. 5. (a)-(b) Spin excitations dispersion along the $[H-1/3, 2/3]$ direction [the blue path in Fig. 1(d)] in planar (a) and axial (b) AFM states. The gray data points with error bars are fitted peak positions of constant-energy curves. The vertical error bars are the energy resolution of the instrument, and the horizontal error bars are the FWHM of the Gauss fittings. The black solid lines are the calculated spin wave dispersion using the LSWT with the parameters in Table I. (c)-(d) High-energy spin excitations measured with $E_i = 400$ meV.

be the lower part of two crossed spin wave branches, with the upper part of the crossed branches becoming weak and diffusive for entering the so-called Stoner continuum. The band crossing-like features can be further supported by the LSWT. In Figs. 5(a) and 5(b), the black solid lines are the calculations from the LSWT using the parameters in Table I. The calculations indeed show band crossings at the vertices of the measured downward cones. Such kind of band crossing is known as the criterion of Dirac magnons [53–57]. This indicates that we may have found the experimental evidence for the existence of Dirac magnon in $\text{Fe}_{0.89}\text{Co}_{0.11}\text{Sn}$. However, the existence of the itinerant Stoner continuum stops us from investigating this feature further. We note that a similar band crossing feature was also observed in pure FeSn and argued to be damped Dirac magnons [38].

Another noteworthy point is that a magnetic flat band from the quasiparticle excitations between the spin-up flat electronic band (majority electrons) and spin-down flat electronic band (minority electrons) has been proposed for 2D FM metals with Kagome lattice [37]. In the case of FeSn, despite the presence of AFM order below T_N , it was treated as a quasi-2D FM metal for the weak AFM coupling between the adjacent FM planes [37, 38]. Starting from such a quasi-2D FM metal, the theoretical calculations predicated a magnetic flat band of the spin excitations in FeSn. However, according to the INS results such a flat band is absent up to ~ 300 meV in FeSn [37, 38]. $\text{Fe}_{0.89}\text{Co}_{0.11}\text{Sn}$ has a similar AFM transition temperature ($T_N \approx 340$ K), same planar AFM order (below $T'_N \approx 70$ K), and comparable ordered magnetic moment [39] with the pure FeSn. Therefore, we would expect that $\text{Fe}_{0.89}\text{Co}_{0.11}\text{Sn}$ has a similar electronic structure and itineracy to FeSn. This means that the predicated magnetic flat band for FeSn [37] is expected to exist in $\text{Fe}_{0.89}\text{Co}_{0.11}\text{Sn}$. Since such a flat band has not been observed up to ~ 210 meV (measurements with $E_i = 300$ meV) in our $\text{Fe}_{0.89}\text{Co}_{0.11}\text{Sn}$ sample, we then measured the higher energy spin excitations with $E_i = 400$ meV to see if there is a magnetic flat band in higher energy regions. We found the strong spin wave dispersion below ~ 100 meV and the weak Stoner continuum intensity up to ~ 250 meV only [Figs. 5(c) and 5(d)]. Our results show that there is no sign of the localized magnetic flat band in $\text{Fe}_{0.89}\text{Co}_{0.11}\text{Sn}$ up to ~ 320 meV. The absence of such a flat band in experiments could have three possible reasons. (i) The flat band is too weak to be visible. (ii) The flat band may mix with the general Stoner continuum from other transition channels and could lose its flatness, narrowness [52]. Together with the possibly low intensity as mentioned in (i), the flat band could become indistinguishable. (iii) The flat band does not exist, that is inconsistent with the theoretical prediction [37].

V. SUMMARY

We have performed systematic neutron scattering measurements on the Kagome-lattice AFM metal $\text{Fe}_{0.89}\text{Co}_{0.11}\text{Sn}$. The planar and axial AFM ordered states are confirmed by neutron diffraction and magnetization results. The careful analyses of the diffraction and the low-energy spin wave results demonstrate that the weak magnetic Bragg peaks at $Q = (0, 0, n + 1/2)$ ($n = \text{integer}$) of the axial state are intrinsic and come from the small in-plane magnetic moment components. Although

464 it has been well confirmed that the ordered moments stack
 465 antiferromagnetically along the c axis, the spin-excitation
 466 spectra are dominated by the in-plane FM spin excitation,
 467 which indicates quasi-2D magnetism. The INS shows a
 468 sharp spin wave below ~ 90 meV, above which the spin
 469 excitations become weak and diffusive, but persist up to
 470 ~ 250 meV. The sharp acoustic spin wave band can be
 471 described in the frame of LSWT by a Heisenberg J_1 - J_2 - J_c
 472 model considering weak single-ion anisotropy. In the axial
 473 AFM state, although the generalized exchange coupling
 474 parameters SJ_1 , SJ_2 , and SJ_c are smaller than that in the
 475 planar state, J_1 and J_2 may show the opposite behavior if
 476 the change of effective spin value is considered. Above the
 477 acoustic magnon, the Stoner continuum appears, which
 478 makes the optical magnons highly damped and invisible.
 479 At the K points of the BZs, we give evidence for the
 480 existence of the Dirac magnon with the upper part of
 481 the Dirac cone becoming weak and decayed in the Stoner
 482 continuum. The magnetic flat band is demonstrated to
 483 be absent in $\text{Fe}_{0.89}\text{Co}_{0.11}\text{Sn}$ up to ~ 320 meV. Our results
 484 give a comprehensive overview of the INS experiments
 485 and LSWT calculations on Kagome-lattice AFM metal
 486 $\text{Fe}_{0.89}\text{Co}_{0.11}\text{Sn}$. The absence of the two optical magnon
 487 branches and the upper part of the Dirac cone highlights
 488 the indispensable role of the itinerant electrons in under-

standing the magnetism in $\text{Fe}_{0.89}\text{Co}_{0.11}\text{Sn}$.

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