

Nematic Order and Orbital Selective Mott State in a Partially Filled Kagome Flat Band

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Coulomb interactions among charge carriers that occupy an electronic flat band (FB) can give rise to captivating phenomena such as quantum criticality, Mott-Hubbard states, and unconventional superconductivity at different FB filling fractions. Consequently, the search for new FB materials with tunable charge carrier filling and strong interactions is a central research theme to broaden our fundamental understanding of strongly interacting quantum systems and to realize materials with novel functionalities. In this work, we present experimental evidence obtained from temperature-dependent scanning tunneling microscopy measurements for various strongly correlated states that appear in partially occupied kagome FBs of $\text{Co}_{1-x}\text{Fe}_x\text{Sn}$ whose filling can be controlled by the Fe-doping level x . At elevated temperatures ($T \geq 16$ K), we find evidence for rotation symmetry breaking across a broad doping range $0.05 < x < 0.25$. Comparison with model calculations reveals that strong Coulomb interactions ($U > 100$ meV) blend the states of two $3d$ -orbital-derived FBs and impart a nematic order parameter. Introducing phase-sensitive lock-in measurements as a local scanning tunneling microscopy probe of incompressible states, we also find direct evidence for Hubbard bands in the partially filled kagome flat bands of samples with ideal Fe doping ($x = 0.17$). Results from Hubbard model calculations suggest the presence of an orbital-selective Mott state in a half-filled kagome flat band. Doping-dependent measurements show that the orbital-selective Mott state descends into a correlated metal phase upon electron and hole doping, marked by a sudden collapse of the spectral gap. Our observations demonstrate that the electronic ground state of the kagome FBs sensitively depends on the complex interplay between geometric frustration, strong Coulomb repulsion, orbital degeneracy, and filling fraction at different temperatures. More broadly, our research establishes kagome lattice materials as a unique platform for novel quantum states that arise from strong electronic correlations in a topologically nontrivial FB.

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I. INTRODUCTION

Strong interactions among electric charge carriers at microscopic length scales can give rise to novel quantum states such as high-temperature superconductivity, non-Fermi liquids, and correlated insulating states significantly. These phenomena influence the macroscopic material properties and can facilitate novel technological applications in

energy-efficient electronic devices, medical imaging, and transportation [1]. From a physical point of view, the appearance of these characteristics is often related to the suppression of the kinetic energy (W) of charge carriers occupying an electronic flat band (FB). As they move more slowly, the effect of the mutual Coulomb interaction U can prevail, and novel electronic ground states emerge at $W < U$.

Prime examples of strongly correlated materials are transition metal oxides [2,3], whose strongly phase diagram arises from strong Coulomb interactions within the spatially localized d orbital of the transition metal ion, as well as the magnetic field-induced FB of quantum Hall systems [4,5]. Recent attention has also been drawn to the flat bands of moiré materials that host a variety of strongly correlated states at different flat band fillings [6–9]. In particular, the combination of nontrivial band topology and quenched kinetic energy in these systems promotes the study of novel quantum states, such as the fractional Chern insulator, that

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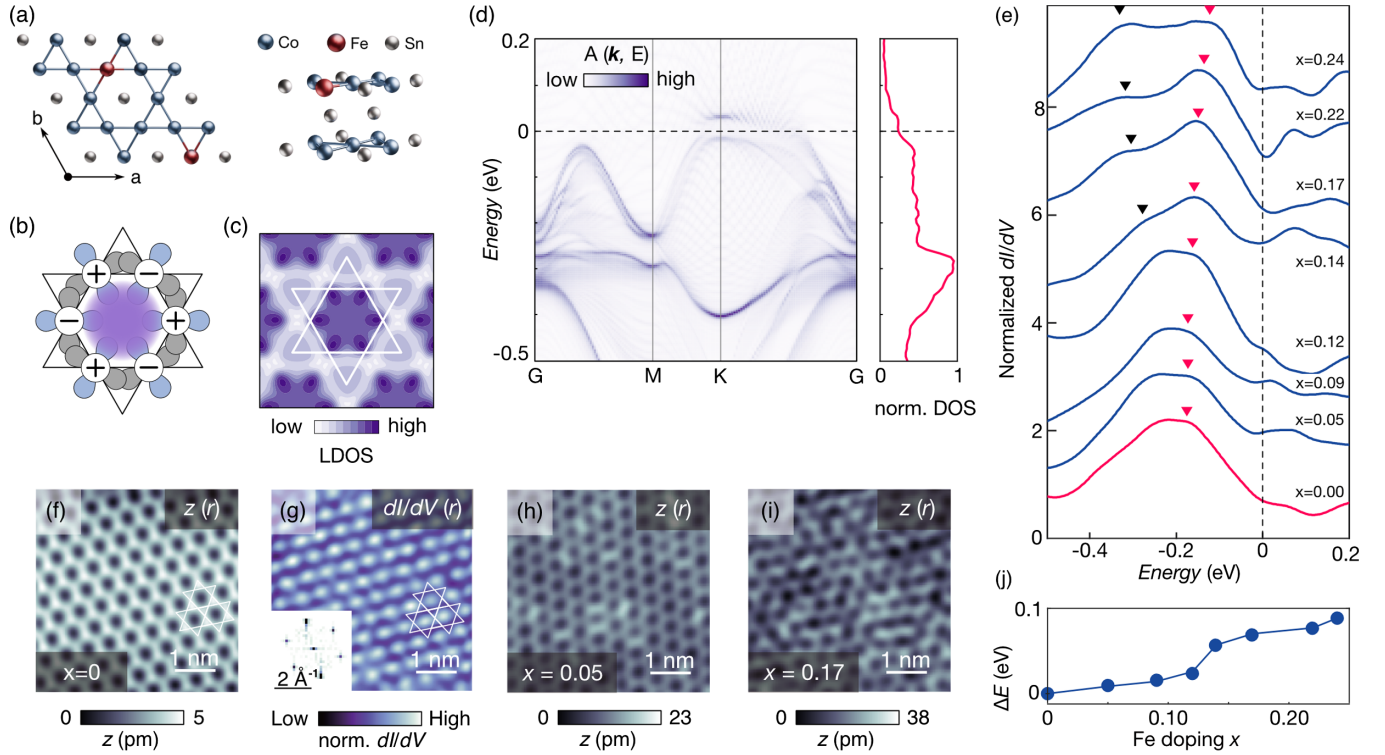


FIG. 1. Realizing partially filled kagome flat bands in Fe-doped CoSn. (a) Schematics of the in-plane lattice structure of $\text{Co}_{1-x}\text{Fe}_x\text{Sn}$ (left) and the layer stacking in the out-of-plane direction (right). Doping with iron (Fe) substitutes cobalt (Co) atoms on the kagome lattice sites. (b) Schematic illustration of the kagome flat band orbital (purple shading) arising from electron localization by destructive interference of neighboring wave functions with opposite phase ($\pm$). (c) Calculated local density of states of the 3d-orbital-derived flat bands of CoSn. (d) Calculated surface spectral function $A(\mathbf{k}, E)$ of CoSn plotted as a function of the in-plane momentum ($\mathbf{k}$) along the high-symmetry lines and the energy E . The right panel shows the calculated DOS. Details of the model calculation are shown in Sec. I in Supplemental Material [29]. (e) dI/dV spectra recorded on the surface of samples with different indicated doping Fe level x at $T = 4$ K. The spectra are vertically offset for clarity ($V = 0.8$ V, $I = 3.0$ nA, $V_m = 10$ mV). The spectra of undoped CoSn ($x = 0$) is highlighted in red color. (f) STM topography recorded on the surface of CoSn ($x = 0$, bias voltage $V = 100$ mV, tunnel current set point $I = 2.0$ nA). (g) dI/dV map recorded at $V = 0$ V ($I = 4.0$ nA, lock-in modulation voltage $V_m = 1.0$ mV) in the same field of view as (f). The inset shows the corresponding 2D-FFT, where dark and bright color correspond to high and low amplitude, respectively. (h), (i) Shown are STM topographies recorded on samples with $x = 0.05$ and 0.17 Fe doping, respectively ($V = 1$ V, $I = 2.0$ nA). (j) Energy shift ΔE of the FB dI/dV peak maximum, indicated by red triangle markers in (e), plotted as a function of Fe doping x .

were inaccessible before [10–13]. This physical richness motivates the search for other materials and concepts toward realizing topologically nontrivial electronic FBs, with the ultimate goal of finding a comparable repertoire of interesting quantum states extending to elevated temperatures.

In the 2D kagome lattice, which consists of corner-sharing triangles [see Fig. 1(a)], destructive quantum interference between electron-wave functions of adjacent lattice sites can give rise to a FB with electronic states that are strongly localized in real space at atomic length scales. When the kagome lattice is composed of transition metal atoms [14–24], the presence of substantial Coulomb interactions ($U \approx 5\text{--}6$ eV) within the d -orbital-derived FB states [17] could promote the emergence of strongly correlated electronics states when the band is partially filled. Recent reports of non-Fermi liquid behavior and anomalous magnetism indicate a measurable effect of electronic interactions in

kagome materials [22,25–28]. However, a fundamental challenge remains to achieve and control partial fillings of the kagome FBs that would allow for the observation of a filling-dependent phase diagram of distinct strongly correlated states.

II. ELECTRONIC FLAT BANDS IN CoSn

CoSn has emerged as a highly promising candidate for realizing correlated phases in electronic flat bands. As seen in Fig. 1(a), cobalt (Co) atoms arrange in a 2D kagome lattice within Co_3Sn planes, and a spacer layer of stanene (Sn_2), composed of tin (Sn), promotes an effective 2D nature of the electronic states [18]. Destructive interference between two sets of Co 3d orbitals residing at the kagome lattice sites, as schematically shown in Fig. 1(b), results in two electronic FBs near Fermi energy E_F . Owing to their 3d-orbital origin, the resulting FB orbitals and their resulting local density of states (LDOS) are localized in

the honeycomb center of the kagome lattice [24], as seen Fig. 1(c). Hence, the spectral characteristics of the CoSn kagome FBs can be directly addressed and distinguished from other electronic states by using scanning tunneling microscopy experiments [24]. It is also worth noting that the FB orbitals are centered inside the honeycomb rings. Correspondingly, the effective real-space descriptions of the FB, also called compact localized states, are distinct from the on-site atomic Co $3d$ orbitals—a characteristic feature of the kagome FB.

The calculated momentum- ($\mathbf{k}$) and energy- (E) dependent surface spectral function $A(\mathbf{k}, E)$ of CoSn is shown in Fig. 1(d) (details of the tight binding model are shown in Sec. I in Supplemental Material [29]). This spectrum, which quantitatively reproduces previous results from angle-resolved photoelectron spectroscopy studies conducted on CoSn bulk samples [17,44], is characterized by two electronic flat bands just below E_F with an in-plane dispersion of the order of $W \approx 100$ –200 meV and a characteristic density of states (DOS) peak at $E \approx -300$ meV. The center of this peak is mostly contributed by electronic states near the Γ and M points of the Brillouin zone, whereas states at higher energy can be attributed to electronic states near the K point. Consistent with this understanding, our results from scanning tunneling spectroscopy measurements conducted on the surface of CoSn, shown in Fig. 1(e) (red solid line), exhibit a pronounced dI/dV peak below the Fermi energy [24,44].

What renders CoSn particularly interesting toward searching for correlated electron phases are two key factors. First, on-site Coulomb repulsion U within the $3d$ -orbital-derived FB states ($U \approx 5$ –6 eV in the unscreened limit) [17] is expected to surpass the kinetic energy of the FB electrons. Second, the $3d_{xy, x^2-y^2}$ -orbital-derived upper FB already touches Fermi energy at the K point of the Brillouin zone in the absence of hole doping [17], as also seen in Fig. 1(d). This understanding is supported by our results from our spectroscopic mapping measurements on CoSn, shown in Figs. 1(e) and 1(f). The dI/dV map, which is recorded at the Fermi energy $E = 0$, visualizes the FB local density of states at the honeycomb center of the kagome lattice [24]. Hence, even a small amount of Fe doping of CoSn could realize substantial filling fractions of the upper FB, favoring the emergence of strongly correlated states in the limit $W/U \sim 1$.

In this work, we report the observation of a rich variety of strongly correlated states in the partially filled kagome FBs of CoSn. Chemical Fe doping of epitaxially grown CoSn thin films lowers the Fermi energy and enables us to realize partial FB occupations. Examining the low-energy electronic states across a wide Fe doping ($0 \leq x \leq 0.24$) and temperature ($4 \text{ K} < T \leq 25 \text{ K}$) range with scanning tunneling microscopy (STM), we discover nematic order, an orbital-selective Mott state, and a correlated metal phase at different temperatures and flat band fillings.

III. REALIZING PARTIALLY FILLED KAGOME FLAT BANDS IN CoSn

We used molecular beam epitaxy (MBE) to realize a doping series of $\text{Co}_{1-x}\text{Fe}_x\text{Sn}$ films (nominal thickness of 50 nm) with different Fe doping levels $0 \leq x \leq 0.24$. Details of the MBE process are described in Ref. [24] and the Appendix; the materials characterization is described in Sec. II in Supplemental Material [29]. The Vollmer-Weber growth mode results in flattop islands of a few hundred nanometers in diameter, whose surface is commonly terminated by a Co_3Sn layer with an apparent honeycomb structure [Fig. 1(c)] [24,45].

Doping of CoSn with Fe manifests in atomic-scale modulations of the topographic height of the Co_3Sn surface layer [Fig. 1(h)] that can be detected only at the lattice sites of the kagome lattice but not inside the honeycomb center. This suggests that the Fe atoms substitute the Co atoms in the Co_3Sn layer and to not take up interstitial lattice sites. The density of these atomic-scale modulations continuously increases with increasing doping levels, as seen in Figs. 1(h) and 1(i). At the same time, we do not find clustering and subphase signatures in the islands studied in the STM measurements, and we can detect large atomically flat terraces in topographic STM measurements on $\text{Co}_{1-x}\text{Fe}_x\text{Sn}$ samples (topographies recorded in samples with other doping levels are presented in Sec. III in Supplemental Material [29]). This suggests the successful formation of $\text{Co}_{1-x}\text{Fe}_x\text{Sn}$ thin films for $0 \leq x \leq 0.24$ [19].

At a spectroscopic level, Fe-doping of CoSn results in a shift of the FB dI/dV peak toward higher energy, as seen in Fig. 1(e) and indicated by red markers. This trend is consistent with the expectation that substituting Co by Fe results in hole doping of the electronic states [19]. A pronounced dI/dV peak can be detected throughout the complete doping series. This suggests that Fe doping induces modest impurity potentials on the order of or weaker than the FB spectral width < 100 meV. Note that we cannot detect spectroscopic signatures of a Kondo effect near Fermi energy at any doping level. This suggests the absence of local magnetic moments of Fe upon their integration into the kagome lattice structure. Moreover, at finite doping levels, the central dI/dV peak of the FB at $E < 0$ develops a spectral splitting, indicated by black markers, and a redistribution of spectral weight to $E > 0$ can be detected. We later show that this splitting is induced by a nematic order parameter that arises at partial FB fillings over a wide doping range. To quantify the energetic shift ΔE of E_F induced by Fe doping, we trace the spectral position of the FB dI/dV peak as a function of x . We detect $\Delta E > 90$ meV at $x = 0.24$, as shown in Fig. 1(j). The energetic shift is particularly apparent at higher doping $x > 0.10$, where E_F is located within the dI/dV peak. Hence, the upper FB is partially filled. In the following, we examine the modifications of the low-energy electronic

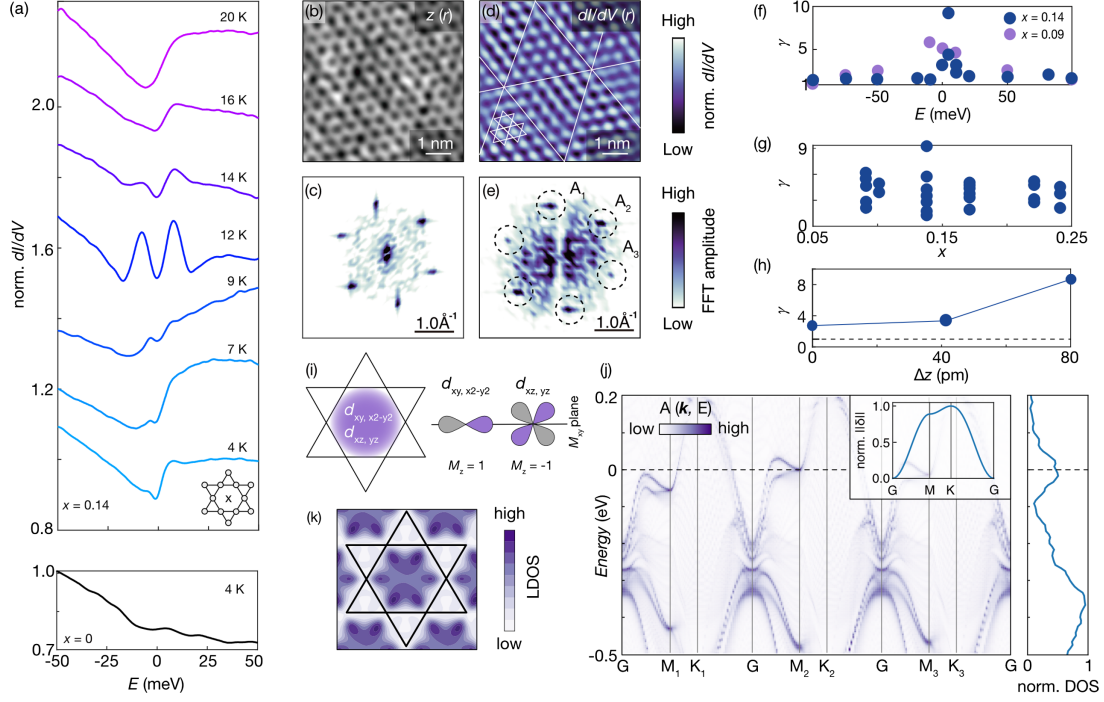


FIG. 2. Nematic order in the partially filled kagome flat bands. (a) Shown are normalized dI/dV spectra recorded at the honeycomb center of the kagome lattice different indicated temperatures T on a sample with $x = 0.14$ Fe doping. The data are vertically offset by 0.2 for clarity, and the STM tip position on the kagome lattice is schematically indicated by a black cross in the inset. The bottom panel shows a corresponding spectrum recorded on undoped ($x = 0$) CoSn ($V = 50$ mV, $I = 3.0$ nA, $V_m = 1.0$ mV). (b), (c) Shown are an STM topography and corresponding 2D-FFT, respectively, recorded on the surface of $\text{Co}_{0.86}\text{Fe}_{0.14}\text{Sn}$ ($V = 100$ mV, $I = 4.0$ nA, $T = 16$ K). (d), (e) Shown are a dI/dV map and corresponding 2D-FFT recorded in the same field of view as (b) ($V = 5$ mV, $I = 4.0$ nA, $V_m = 1.0$ mV, $T = 16$ K, $\Delta z = 40$ pm). The dI/dV map is a Fourier-filtered inverse FFT of the Bragg peaks [black circles in (e)] to remove low-frequency noise stemming from electronic background variations induced by the Fe doping. Raw data are shown in Sec. IV in Supplemental Material [29]. (f)–(h) Shown are the dependence of the symmetry-breaking strength γ on the energy $E = eV$ (e , electron charge), the Fe doping x , and the reduction Δz of the tip-sample separation. y -axis error bars are included within the symbols. (i) Schematic illustration of the spatial position of the flat band orbitals derived from Co $3d_{xy/x^2-y^2}$ and $3d_{xz/yz}$ orbitals (left). The mirror eigenvalues of these orbitals with respect to the mirror- z axis are indicated (right). (j) Calculated surface spectral function $A(\mathbf{k}, E)$ of CoSn in the presence of a finite nematic order parameter $\delta = 200$ meV plotted as a function of the in-plane momentum ($\mathbf{k}$) along the high-symmetry lines and the energy E . The right panel shows the calculated DOS. The inset displays the calculated norm of the matrix of the mean-field order parameter δ normalized to its maximum value at the crystallographic K point along the high-symmetry lines of the Brillouin zone. (k) Shown is the calculated local density of states at the surface of a 50 unit cell slab of CoSn with an interband order parameter $\delta = 180$ meV. See Sec. VI in Supplemental Material for calculation details [29].

structure near E_F of the partially filled FBs of $\text{Co}_{1-x}\text{Fe}_x\text{Sn}$ as a function of temperature and iron doping.

In Fig. 2(a), we show dI/dV spectra recorded at the honeycomb center of the kagome lattice of moderately doped $\text{Co}_{0.86}\text{Fe}_{0.14}\text{Sn}$ when the temperature increases from 4 to 20 K. At all temperatures, the dI/dV spectra of the $x = 0.14$ -doped sample exhibit a low-energy structure at Fermi energy that can be distinguished from the featureless dI/dV spectrum recorded on undoped ($x = 0$) CoSn. At 4 K, our experimental base temperature, the sample exhibits a pronounced and electron-hole asymmetric dip-peak feature at E_F . As the temperature increases, the dI/dV spectrum further undergoes two transitions; at $T \approx 8$ K, we observe a redistribution of the spectral weight into an approximately electron-hole symmetric double peak feature; at 16 K, the spectral weight is redistributed again into

an electron-hole asymmetric dI/dV spectrum. These observations suggest the presence of a temperature-driven cascade of low-energy electronic states in the partially filled FB of Fe-doped CoSn.

IV. OBSERVATION OF ROTATIONAL SYMMETRY-BREAKING DENSITY OF STATES

To shed light on the origin of these low-energy density of state modifications, we first examine the electronic properties of $x = 0.14$ -doped sample at $T \geq 16$ K. We have performed spectroscopic mapping experiments to visualize the real-space distribution of the dI/dV amplitude associated with the electron-hole asymmetric feature at E_F , as seen in Fig. 2(a). In Figs. 2(b)–2(e), we show the resulting STM topography and dI/dV map recorded in the same

field of view, as well as the corresponding 2D fast Fourier transform (2D-FFT) maps. The STM topography is characterized by the apparent honeycomb structure of the kagome lattice in STM measurements [24] and topographic height modulation owing to the presence of Fe dopants. The 2D-FFT exhibits six Bragg peaks detected of the same amplitude, mirroring the sixfold rotational symmetry of the kagome lattice. In contrast, the dI/dV map recorded at $E = 5$ meV lacks the uniform appearance, previously detected at the surface of undoped CoSn [cf. Fig. 1(g)] and exhibits a patchwork of domains that encompass an area of several tens of unit cells. The domain boundaries are indicated by white lines. In some of these domains, the dI/dV amplitude appears as elongated stripes, a clear deviation from the pointlike appearance of the local flat band density of states detected in undoped CoSn shown in Fig. 1(g). This observation also manifests itself in the corresponding 2D-FFT map shown in Fig. 2(e), in which two out of six Bragg peaks (highlighted by black dashed circles) exhibit a suppressed amplitude. A closer inspection shows that the six Bragg peaks can be categorized into three subsets (A_1 , A_2 , and A_3) of pairs that have different relative amplitudes ($A_1 = 0.9$, $A_2 = 1.0$, and $A_3 = 0.1$). For simplicity, we parametrize the strength of C_{6z} rotation symmetry breaking as $\gamma = (A_1 + A_2)/(2A_3)$; C_{6z} is broken for $\gamma \neq 1$. We find $\gamma = 9.5$ for the dI/dV map shown in Fig. 2(d). This value is in stark contrast with the dI/dV maps recorded on undoped CoSn ($\gamma = 1.6$), as shown in Fig. 1(f), which is uniform across the field of view and does not exhibit domains.

Note that an anisotropic tip shape and strain could generally result in an apparent rotation symmetry breaking with small deviations of γ from unity [46]. However, the presence of domains in the dI/dV spectra recorded near E_F unambiguously rules out these effects as the primary cause of the observed C_{6z} -symmetry breaking. By comparison, the corresponding topography recorded in the same field of view, shown in Fig. 2(b), does not exhibit crystallographic domains. Moreover, the periodic density of states modulation is uniform across each domain and gives rise to distinct Bragg peaks in the 2D FFT. Therefore, these domains cannot also be caused by randomly distributed local defects that were found to induce C_{6z} -symmetry-breaking orbital states in the isostructural FeSn [47]. The charge density wave can also be ruled out, as the discrete translation symmetry is preserved even though the rotation symmetry C_{6z} is broken. Therefore, our combined observations suggest that electronic correlations induce a nematic state in the low-energy electronic structure that reduces the sixfold rotation symmetry (C_{6z}) of the kagome lattice to a twofold (C_{2z}) rotation symmetry.

To obtain more experimental insight into the origin of this state, we examined the rotational symmetry breaking as a function of several experimental parameters, as shown in Figs. 2(f)–2(h). First, we have examined dI/dV maps recorded at different energies away from E_F . As can be seen

in Fig. 2(f), we detect signatures of C_{6z} breaking over a wide energy range from $-100 \text{ meV} \leq E \leq 100 \text{ meV}$, and this effect is most pronounced ($\gamma = 9.5$) near Fermi energy (energy-dependent dI/dV maps are displayed in Sec. IV A in Supplemental Material [29]). Next, we studied the effect of the Fe doping level on the breaking of C_{6z} . We detect rotation symmetry breaking in all samples with $x \geq 0.09$ as shown in Fig. 2(g) (also see Sec. IV B in Supplemental Material [29]). Only in weakly doped samples at $x = 0.05$ can we not detect C_{6z} to C_{2z} breaking as shown in Sec. V in Supplemental Material [29]. We also find that the size of the domains in the low-energy electronic structure increases, when the Fe doping level is reduced, suggesting that impurity potentials induced by the Fe dopants act as pinning centers, as shown in Sec. IV B in Supplemental Material [29]. Finally, we examined the response of the symmetry breaking to a variation in the tip-sample separation. This measurement is valuable, because the in-plane crystallographic momentum of the charge carriers modulates the effective height of tunnel barrier between the tip and sample (see Sec. IV C in Supplemental Material for a detailed discussion of this effect [29]) [48]. We find that C_{6z} breaking increases when decreasing the tip-sample separation by Δz [as seen in Fig. 2(h)]. This suggests that symmetry breaking is more prominent in electronic states with finite in-plane momentum, which have a larger out-of-plane wave function decay constant than those states near the crystallographic Γ point.

V. NEMATIC ORDER IN A PARTIALLY FILLED KAGOME FLAT BAND

Given the localized character ($\approx 2\text{--}3 \text{ \AA}$) of the kagome FB orbitals, it is natural to consider a symmetry-breaking order parameter built from the two $3d_{xy/x^2-y^2}$ and $3d_{xz/yz}$ FB orbitals localized at the honeycomb center of the kagome lattice [24], as schematically illustrated in Fig. 2(i). Microscopically, such a phenomenological model is justified given the strong density-density interaction in the $3d$ orbitals, which is on the order of $U \approx 5\text{--}6 \text{ eV}$ in the unscreened limit [17]. On phenomenological grounds, we cannot rule out the possibility of alternative microscopic origins, say, electron-phonon coupling, for the nematic state. However, to account for the observation of C_{6z} -symmetry breaking over the large energy window of $\pm 100 \text{ meV}$ from the Fermi energy, an order parameter on the same order of magnitude is required and that suggests that system is far from the weak-coupling limit. If only one of the two FBs is involved, the mean-field (MF) symmetry-breaking Hamiltonian can be expressed as $\delta\hat{H}_{\text{MF}} = \sum_{\vec{r},\mu,\nu} \hat{f}_{\vec{r},\mu}^\dagger (\vec{d} \cdot \vec{\sigma})_{\mu\nu} \hat{f}_{\vec{r},\nu}$, where $\hat{f}_{\vec{r},\mu}^\dagger$ creates a localized electron centered around $\vec{r}$ with spin $\mu = \uparrow, \downarrow$, $\vec{d}$ is a three-dimensional real vector that parametrizes the symmetry-breaking order parameter, and $\vec{\sigma}$ denote the three Pauli matrices. This order parameter transforms as a spin one and always breaks time-reversal symmetry. If $\vec{d}$ points along the z

direction, C_{6z} is preserved; if $\vec{d}$ lies in the plane, no rotation symmetry remains. Neither of these scenarios is compatible with our experimental observations.

We are, thus, naturally led to considering interband interactions between states belonging to the two FBs. First, note that the in-plane orbitals $3d_{xy/x^2-y^2}$ are even under a mirror M_{xy} , which flips $z \leftrightarrow -z$, whereas the out-of-plane orbitals $d_{xz/yz}$ are odd under the same mirror, as shown in Fig. 2(i). Therefore, these two sets of kagome FBs can hybridize only when spin-orbit coupling is present. The spin-orbit coupling strength controls the degree of hybridization between these two orbitals, which is rather weak in the noninteracting limit. Since the energetic separation between the FB states is only on the order of a hundred millielectron volts or less [17], a screened Coulomb interaction $U' \approx 100$ meV could induce an interband order parameter $\delta \propto U$ that mixes the electronic states of the two FBs.

Our symmetry analysis identifies exactly one MF interband order parameter δ which reduces C_{6z} to C_{2z} (see Sec. VI in Supplemental Material [29]). This order parameter hybridizes the states of the upper and lower FBs over a wide energy range $\propto \delta$ and results in a ground state with only C_{2z} symmetry, as seen from the calculated energy- and momentum-dependent spectral function and local density of states shown in Figs. 2(j) and 2(k). Mixing of the two FBs is especially prominent for states with large in-plane momentum near the K and M points but vanishes near the G point of the Brillouin zone, as illustrated by the momentum-dependent norm of the order parameter matrix [see inset in Fig. 2(j)]. This effect manifests in the spectral splitting of the calculated FB density of state peak, as shown in the right in Fig. 2(j). Here, a new FB DOS peak at Fermi energy arises from the Van Hove singularity at the M point of the upper FB, and the DOS peak below Fermi energy contains additional spectral weight from the Van Hove singularity at the M point of the lower FB that is pushed down to approximately -400 meV. This effect of the introduced order parameter is in agreement with our experimental observation of a spectral splitting of the central FB dI/dV peak at $E < 0$ and additional spectral weight arising above Fermi energy at finite Fe doping, as seen in Fig. 1(e). Moreover, the alignment of the M point of the mixed FB with Fermi energy could also account for the particularly strong C_{6z} breaking detected near Fermi energy [see Figs. 2(d)–2(f)].

Finally, we have conducted self-consistent Hartree-Fock calculations of an extended Hubbard model on the kagome lattice to identify the energetically most favorable ground state of the partially filled kagome flat band in the presence of on-site U and intersublattice repulsions [49]. Although the calculations are done for a simpler single-orbital kagome model, our results show that electronic nematicity is a generic outcome of partially filled kagome flat bands over a wide filling and interaction

range. Interestingly, while pure on-site repulsion would favor a ferromagnetic ground state, intersublattice interactions are a critical ingredient to stabilize a nematic ground state. Such sizable intersublattice interaction is naturally anticipated due to the characteristic charge distribution of the FB states around the hexagon rings and has been reported in numerical calculations estimating the interaction parameters in kagome compounds [45]. In a multiorbital kagome system, like Fe-doped CoSn, where the energetic separation between the FBs is comparable to the interaction strength, we expect the mentioned mechanism to lead to an interband nematic order parameter as is consistent with our phenomenological analysis. Overall, our theoretical analyses show that an interband nematic order parameter, arising from strong electron-electron interactions, provides a natural explanation for the spectral modifications of the FB peak detected in dI/dV spectra [Fig. 1(e)] and the rotational C_{6z} to C_{2z} symmetry breaking detected in our dI/dV maps [Fig. 2(d)], and it can also account for all salient experimental characteristics, such as the observed energy, doping, and tip-height dependence [Figs. 2(f)–2(h)].

VI. OBSERVATION OF DOPING-DEPENDENT SPECTRAL GAPS AT PARTIAL FLAT BAND FILLING

Next, we focus on the double peak feature observed in the dI/dV spectrum near Fermi energy between 8 and 16 K, as seen in Fig. 2(a), and examine its response to changes in the Fe doping. In Fig. 3(a), we display representative dI/dV spectra recorded at the kagome honeycomb center of the kagome lattice on the surface of samples with different Fe doping x at $T = 12$ K; dI/dV spectra recorded on samples with other intermediate doping levels are shown in Sec. VII A in Supplemental Material [29]. We find that double peaks near E_F can be detected across the complete examined doping sequence $0.09 \leq x \leq 0.24$. Only at small doping level $x = 0.05$ is this spectral feature absent, as shown in Sec. V in Supplemental Material [29]. At intermediate doping ($x = 0.17$), we further observe a qualitatively different dI/dV spectrum [red line in Fig. 3(a)]. It is characterized by two pronounced peaks detected at $E \approx -40$ meV and $E \approx 70$ meV, respectively. The two peaks are separated by a large flat gap $E_G \approx 110$ meV, which is asymmetric about E_F and appears on top of a finite dI/dV background. The doping evolution of E_G , shown in Fig. 3(b), highlights that this wide-gap feature can be detected only in samples with $x = 0.17$ and that the gap size renormalizes rapidly to approximately 20 meV upon doping with electrons and holes.

We have mapped out the spectral weight distribution of the detected dI/dV peaks in real space for samples with different Fe doping. dI/dV maps recorded at the peak energies above E_F on the surface of samples with $x = 0.14$

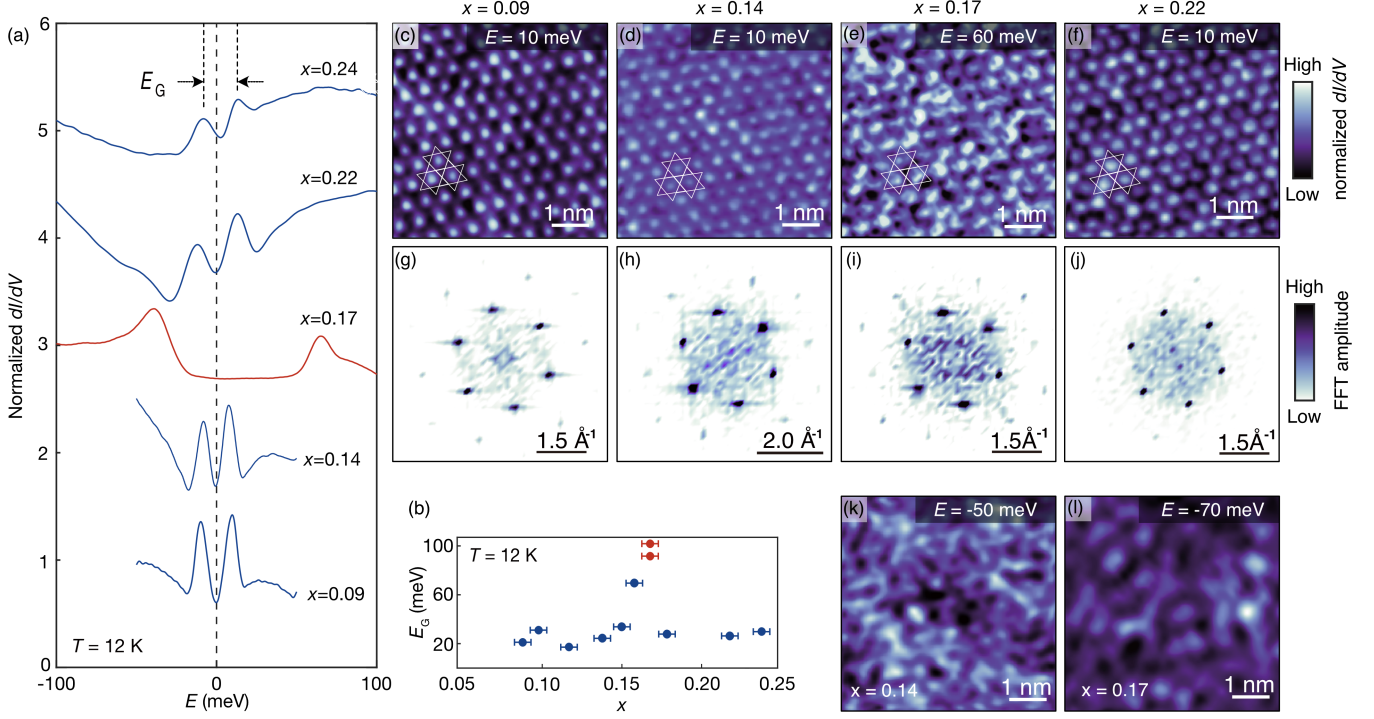


FIG. 3. Observation of a doping-dependent spectral gap at partial flat band fillings. (a) Normalized dI/dV spectra recorded on $\text{Co}_{1-x}\text{Fe}_x\text{Sn}$ samples with different indicated doping levels x ($V = 150$ mV, $I = 4.0$ nA, $V_m = 1.0$ mV, $T = 12$ K). The spectra are vertically offset by unity for clarity. (b) Spectral separation E_G of the double peaks appearing in the dI/dV spectra of (a), plotted as a function of x . y-axis error bars are included within the symbols. (c)–(f) Spatial distribution of the amplitude of the dI/dV peak observed at $V > 0$ in (a) recorded on samples with indicated doping levels $I = 4.0$ nA, $V_m = 1.0$ mV, and $T = 12$ K). (g), (h) 2D-FFT maps of the data shown in (c)–(f). (k), (l) Spatial distribution of the dI/dV amplitude observed at indicated energies outside the double peak structure in (a) recorded on samples with $x = 0.14$ and $x = 0.17$ Fe doping, respectively ($I = 4.0$ nA, $V_m = 1.0$ mV, $T = 12$ K).

and $x = 0.17$ Fe doping are shown in Figs. 3(c)–3(f); the corresponding topographies recorded in the same field of view are shown in Sec. VII B in Supplemental Material [29]. We find that the spectral weight of these dI/dV peaks is strongly localized at the honeycomb center of the kagome lattice at all doping levels. The dI/dV signal forms a periodic real-space pattern that is commensurate with the lattice constant of the kagome lattice. We also find that C_{6z} symmetry breaking of the local density of states is strongly reduced, as can be seen from the 2D-FFT maps shown in Figs. 3(g)–3(j) (also see Sec. VII in Supplemental Material [29]). On the other hand, dI/dV maps recorded further away from Fermi energy lack this real-space periodic dI/dV pattern, as seen in Figs. 3(k) and 3(l). This suggests that the modifications to the low-energy density of states arise from electronic interaction effects.

The dI/dV spectra and dI/dV maps recorded at 12 K are distinct from the characteristics detected at $T \geq 16$ K. This suggests the presence of a distinct electronic ground state. Moreover, our measurements show that the low-energy electronic structure at 12 K is very sensitive to the doping level, because dI/dV spectra recorded on samples with 17% Fe doping can be qualitatively distinguished

from those recorded on samples with other doping levels, as seen in Fig. 3(a). A closer inspection of the spatial distribution of the spectral weight of this peak in Fig. 3(e) reveals an open circular shape (“bitten donut”), which contrasts with the rather dotlike shape of the dI/dV pattern found at other doping levels $x < 0.17$ and $x > 0.17$. In the following, we focus on examining the origin of this state in more detail.

VII. EXPERIMENTAL DETECTION OF AN INCOMPRESSIBLE STATE IN THE KAGOME FLAT BAND ORBITAL

First, we studied the response of the wide-gap feature detected in dI/dV spectra recorded on the $x = 0.17$ sample to a temperature change. Figure 4(a) displays the dI/dV spectra recorded at the indicated temperatures. At temperatures between 4 and 12 K, the dI/dV spectrum remains qualitatively unchanged. When the temperature increases to above 12 K, the dI/dV amplitude of the two peaks decreases and the gap softens. However, even at 24 K, the highest temperature that we can experimentally reach, the two peaks are still visible. Hence, the temperature dependence of the low-energy electronic states of $\text{Co}_{0.83}\text{Fe}_{0.17}\text{Sn}$ is

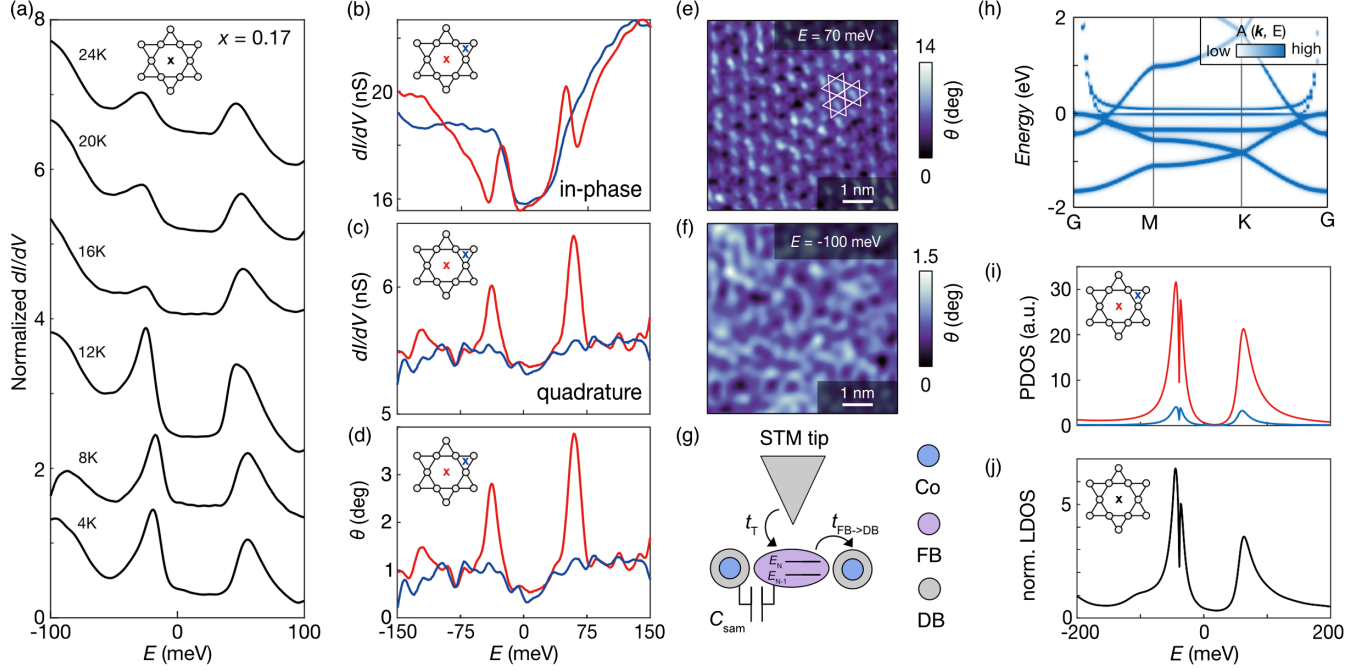


FIG. 4. Experimental evidence for an orbital-selective Mott state in $\text{Co}_{0.83}\text{Fe}_{0.17}\text{Sn}$. (a) Series of normalized dI/dV spectra recorded on $\text{Co}_{0.83}\text{Fe}_{0.17}\text{Sn}$ at different indicated temperatures ($V = 150$ mV, $I = 3.0$ nA, $V_m = 1.0$ mV). The spectra for $T \geq 8$ K are vertically offset for clarity. (b),(c) Shown are the in-phase and quadrature (out-of-phase) components of dI/dV spectra recorded at the honeycomb center and rim positions of the kagome lattice ($V = 150$ mV, $I = 3.0$ nA, $V_m = 1.0$ mV, $T = 12$ K). The STM tip position is schematically indicated in the inset. (d) Lock-in phase θ calculated from the data shown in (b) and (c). (e), (f) Shown are the spatially resolved lock-in phases calculated from the in-phase and quadrature components of the measured dI/dV amplitude at $E = 70$ meV and $E = -100$ meV, respectively. The corresponding in-phase and quadrature maps are shown in Sec. IX in Supplemental Material [29]. (g) Schematic model of the $3d$ -orbital-derived FB (pink color) and dispersive band (DB, gray color) orbitals on the kagome lattice of Co atoms (blue color). The charge states E_N and E_{N-1} of the Hubbard bands can be effectively seen as a quantum dot with sample self-capacitance C_{sam} where the hopping terms t_T and $t_{\text{FB} \rightarrow \text{DB}}$ describe the charge transfer to the STM tip and adjacent Co $3d$ orbitals, respectively. (h) Shown is the calculated surface spectral function $A(\mathbf{k}, E)$ of the Hubbard + I model plotted as a function of the in-plane momentum ($\mathbf{k}$) along the high-symmetry lines and the energy E . (i) Shown is the projected density of states of $A(\mathbf{k}, E)$ at the honeycomb center and rim position of the kagome lattice. (j) Shown is the normalized calculated LDOS at the honeycomb center of the kagome lattice. Details of the Hubbard + I model calculation are shown in Sec. X in Supplemental Material [29].

qualitatively different from those at other doping levels, as exemplified for $\text{Co}_{0.86}\text{Fe}_{0.14}\text{Sn}$ in Fig. 2(a). These observations suggest the presence of a distinct electronic ground state in $\text{Co}_{0.83}\text{Fe}_{0.17}\text{Sn}$ across a wide temperature range.

In Fig. 4(b), we present dI/dV spectra recorded on another CoSn island of the $x = 0.17$ sample, which exhibits characteristics similar to those shown in Fig. 3. Examining the spectral characteristics as a function of the STM tip position, we find that the peaks flanking the wide gap can be detected at the honeycomb center of the kagome lattice only when tunneling into the FB orbitals. However, when the STM tip is located at the rim position of the kagome lattice (see the inset for schematic illustration of tip position), the peaks are suppressed. Moreover, the spectral gap detected in $\text{Co}_{0.83}\text{Fe}_{0.17}\text{Sn}$ exists on top of a finite density of states background, as seen in Fig. 3(a). These observations suggest the presence of an orbital-selective insulating state in the partially filled kagome FB that gaps out a subset of the electronic states at Fermi energy.

To experimentally support this hypothesis, we implement a new methodology with the STM, phase-sensitive lock-in measurements, to effectively probe the compressibility of electronic states at the atomic scale. The key idea is the understanding that tunneling into a locally incompressible state is, in simple terms, equivalent to adding a finite self-capacitance of the sample to the electric circuit, and such a change in the circuit impedance can be detected using lock-in measurements. We describe details of this methodology in Sec. IX in Supplemental Material [29]. To illustrate this concept, we plot the quadrature (out-of-phase) component of the dI/dV spectrum, which is shown in Figs. 4(b) and 4(c). For tunneling into metallic states, we would expect an energy- and tip-position-independent background signal. In contrast, we detect a change in the quadrature amplitude when the tip is located at the honeycomb center and when tunneling occurs into the dI/dV peaks of the wide-gap feature; when tunneling occurs at other energies or spatial positions, the quadrature

remains unchanged. This quadrature change maps to a change in the lock-in phase $\theta = \text{atan}[(Y - Y_0)/X]$ as seen in Fig. 4(d). Here, X , Y , and $Y_0 \approx 5.5$ nS denote the in-phase, quadrature, and quadrature background, respectively. Note that, since we detect a change of the quadrature component, the phase change does not result from energy- and tip-position-dependent changes of the tunnel current that affect the amplitude of the in-phase component. Mapping out this phase change in real space, we find that the phase map recorded at the dI/dV peak energy ($E = 70$ meV) exhibits a periodic real-space variation. As seen in Fig. 4(e), we detect significant phase changes (maximum amplitude $\theta \approx 14^\circ$) that are strongest at the honeycomb center of the kagome lattice while the phase remains unchanged at the rim position. On the other hand, we do not detect a spatially periodic phase change at energies away from the wide-gap feature ($E = -100$ meV) as shown in Fig. 4(f) (the corresponding in-phase and quadrature maps are shown in Sec. IX in Supplemental Material [29]).

The observation of a locally incompressible state, which adds a finite self-capacitance to the circuit, can be naturally explained by considering the presence of Hubbard bands in the partially filled kagome FB. The energy difference between the two relevant charge states of the Hubbard bands is $E_N - E_{N-1} \approx U(N-1) - \mu + eV$ (N and μ denote the charge occupation and chemical potential, respectively). C_{sam} of such discrete energy states vanishes for general values of V but forms a delta function for special values of V , which causes different charging states to be degenerate ($E_N = E_{N-1}$) or resonating. Hence, tunneling between the STM tip and the Hubbard bands will lead to a change of the lock-in phase induced by C_{sam} (as modeled in detail in Sec. IX in Supplemental Material [29]). More generally, one can consider the local Hubbard states in the FB orbital as an effective quantum dot, as shown in Fig. 4(g), where the STM tip and adjacent $3d$ Co orbitals correspond to the leads with hopping amplitudes t_T and $t_{\text{FB} \rightarrow \text{DB}}$, respectively, and $t_{\text{FB} \rightarrow \text{DB}}$ facilitates draining of the charge from the Hubbard bands into the metallic continuum. Note that the quantitative analysis of the detected phase change in terms of an effective capacitance cannot be performed within a simple perturbative framework because of the nonlinearity of the low-energy electronic density of states of Hubbard bands. Further work will be required to analyze this effect using a full Green's function treatment to account for the charge dynamics [50].

VIII. ORBITAL-SELECTIVE MOTT STATE IN A PARTIALLY FILLED KAGOME FLAT BAND

The observation of the energy- and position-dependent change of the lock-in phase provides experimental evidence for the presence of Hubbard bands. We argue that this observation can be interpreted in terms of an orbital-selective Mott (OSM) state [51–53] that exists in the upper,

partially filled kagome FB of $\text{Co}_{0.83}\text{Fe}_{0.17}\text{Sn}$. This state is favored by strong on-site repulsion $U = E_G \approx 120$ meV, where E_G corresponds to the spectral gap in the dI/dV spectrum [cf. Fig. 3(b)] that is on the order of the experimentally detected band width $W \approx 100$ meV such that $U/W \approx 1$ [44,54]. To validate this interpretation on theoretical grounds, we have performed additional model calculations of the low-energy electronic states of the partially filled kagome FB using the Hubbard + I approximation. Details of these model calculations are shown in Sec. X in Supplemental Material [29].

Near Fermi energy, the band structure of CoSn within the two-dimensional Brillouin zone is composed of two $3d_{xz/yz}$ - and $3d_{xy/x^2-y^2}$ -orbital-derived flat bands and two $3d_{xz/yz}$ -derived dispersive bands, whereas the p orbital states of Sn are gapped out to high and low energy via the selective coupling to a subset of the Co $3d$ orbitals [24,45]. Hence, in our model, we approximate the low-energy electronic structure by considering bands contributed by the Co $3d_{xz/yz}$ and $3d_{xy/x^2-y^2}$ orbital. This approximation is justified, because it was previously shown that the two kagome flat bands exhibit quasi-2D characteristics [17,24]. The effect of on-site Coulomb repulsion between the FB electrons is included by an effective Hubbard term $U = 120$ meV, as extracted from the width of the spectral gap in the dI/dV spectrum in Fig. 3(a), and we fix the filling factor of the upper FB near half filling.

This model can be solved self-consistently, and the resulting spectral function $A(\mathbf{k}, E)$ along the high-symmetry lines is shown in Fig. 4(h). As can be seen, the main effect of the presence of on-site electron repulsion is the splitting of the d_{xy/x^2-y^2} -derived FB into upper and lower Hubbard bands, while the dispersive bands remain uncorrelated. On the other hand, the $d_{xz/yz}$ -orbital-derived lower FB, which is nearly fully occupied, shows negligible correlation effects. The projection of $A(\mathbf{k}, E)$ onto the honeycomb center and rim of the kagome lattice is shown in Fig. 4(i). The resulting projected density of states shows that the Hubbard bands are predominantly localized to the FB orbital at the honeycomb center, whereas the rim state carry little spectral weight. Finally, we also calculate the LDOS of $A(\mathbf{k}, E)$ at the honeycomb center, as shown in Fig. 4(j). The calculated LDOS exhibits pronounced Hubbard bands, which are separated by a wide gap and appear on top of a finite density of states, which is contributed by the dispersive $3d_{xz/yz}$ -derived bands. Hence, the results of our model calculations accurately reproduce our experimental observation of a wide-gap feature in dI/dV spectra as well as the spatial distribution of the associated spectral weight, seen in Figs. 4(a) and 4(b), indicating the presence of an OSM state in the partially filled upper kagome FB of $\text{Co}_{0.83}\text{Fe}_{0.17}\text{Sn}$.

IX. DISCUSSION

The OSM state detected at $x = 0.17$ doping does not occur in a stoichiometric compound. This poses challenges

in determining the exact electron filling of the upper FB in our STM experiments. In a simplistic rigid-band model (as shown in Supplemental Material, Sec. I [29]), the Fermi level is expected to shift by $\Delta E' \approx 40$ meV at $x = 0.17$ compared to undoped CoSn. This shift is similar in magnitude to the experimentally detected shift of the FB dI/dV peak maximum approximately 70 meV [cf. Fig. 1(h)] that is predominantly composed of states near the Γ point in the Brillouin zone, as seen in Fig. 1(d). However, the nematic order parameter $\delta \propto U$ increases the energy of the upper FB states, especially near the K and M points, as shown in Fig. 2(j). Therefore, this mechanism is expected to amplify the effective hole doping in this FB beyond the level expected to result from chemical Fe doping within a rigid band model. In light of experimental and theoretical evidence for Hubbard bands obtained through phase-sensitive lock-in measurements and model calculations, we infer that half filling of the upper kagome FB is effectively achieved at $x = 0.17$ Fe doping.

Understanding the strongly correlated ground state that arises from doping a Mott insulator is a notoriously challenging task [2]. In our study, electron ($x < 0.17$) and hole ($x > 0.17$) doping of the OSM state detected in $\text{Co}_{0.83}\text{Fe}_{0.17}\text{Sn}$ results in a sudden collapse of the wide spectral gap into a pair of dI/dV peaks flanking a narrow V-shaped gap, as seen in Fig. 3(a). This transition is accompanied by a significant reduction of the detected lock-in phase change, as shown in Sec. IX in Supplemental Material [29], suggesting that the incompressible nature of the OSM state is strongly reduced but remains finite. These observations indicate that electron-electron interactions alter the low-energy electronic structure even away from ideal filling at $x = 0.17$. Therefore, we coin this doped OSM state a correlated metal state.

In our study, the real-space localized spectral weight detected in samples with $x > 0.17$ and $x < 0.17$ Fe doping does not break translation symmetry, as seen in Figs. 3(c), 3(d), and 3(f). Hence, this phase cannot be explained by a simple charge density wave. Electronic correlations in the presence of disorder can generally result in a logarithmic suppression of the density of states at Fermi energy [55]. However, this mechanism is incompatible with the presence of two spectral peaks and a periodic real-space pattern of the associated spectral weight seen in our measurements presented in Figs. 3(a)–3(f). As shown in Sec. XI in Supplemental Material [29], measurements conducted at the temperature-driven transition into the correlated metal phase indicate the presence of significant charge fluctuations at the transition points; furthermore, the double peak spectrum, as seen in Fig. 3(a), also does not respond to the application of a magnetic field. These combined observations provide a preliminary understanding that the correlated metal phase detected at $x > 0.17$ and $x < 0.17$ is coupled to the charge degree of freedom.

Results of previous model calculations suggest that an OSM state could be separated from the metallic side of the spectrum by a quantum critical point [56]. To date, an OSM state has so far been discussed only in the context of multiband transition metal oxides [51] and iron chalcogenides [57], and its response to electron and hole doping remains unexplored. The kagome FBs of $\text{Co}_{1-x}\text{Fe}_x\text{Sn}$ with the ability to continuously tune the flat band filling fraction could provide new avenues to shed light on the nature of this state and to examine the appearance of quantum criticality and associated superconducting phases [58] at the transition into a correlated metal phase, for example, using Hall and quantum oscillation measurements [53] as well as spectroscopic mapping with the STM [59].

X. CONCLUSION AND OUTLOOK

Overall, our study establishes the presence of various strongly correlated states in the kagome FB of Fe-doped CoSn at different temperatures and filling fractions. Combining thin film growth with STM measurements and theoretical model calculations, we provide conclusive evidence for the presence of nematic order over a wide doping range and an OSM state at $x = 0.17$ Fe doping. The experimental characteristics of these states originate from the $3d$ orbital degeneracy of Co in combination with strong electron-electron interactions at $U \approx W$ in the FB orbitals. This establishes the FBs of CoSn as a distinct electronic system that interpolates between the strongly correlated nature of transition metal oxides [2,3] and the topologically nontrivial character of moiré materials [60,61]. Through these observations, our work facilitates exciting prospects for further exploration of these and other correlated electronic states arising in the partially filled flat bands of Fe-doped CoSn and related compounds [26], efforts that will benefit from reduced disorder in modulation-doped thin films [62] and studies at even higher and lower temperatures. For example, the correlated metal phase arising from doping the OSM state with electrons and holes remains less understood, and it would also be interesting to use spin-sensitive STM probes to shed light on the origin of another nematic low-temperature phase detected at $T \leq 8$ K (see Sec. XII in Supplemental Material [29]) [63]. More broadly, our work opens a new research direction toward discovering novel quantum states in kagome flat bands that emerge from the unique interplay among geometric frustration, orbital degeneracy, and strong electronic interactions at different filling fractions.

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DATA AVAILABILITY

The data that support the findings of this article are openly available [64].

APPENDIX: METHODS

1. Molecular beam epitaxy of CoFeSn thin films

The $\text{Co}_{1-x}\text{Fe}_x\text{Sn}$ thin films presented in this study were prepared using a home-built molecular beam epitaxy system. Details of the film deposition of CoSn films and substrate preparation procedures are described elsewhere [24]. Films of nominal thickness 50 nm were deposited on Nb-doped $\text{SrTiO}_3(111)$ single crystals (CrysTec; dimensions $5 \times 5 \times 0.5 \text{ mm}^3$; Nb concentration 0.05 wt%) by coevaporating Co, Fe, and Sn from thermal effusion cells. The beam equivalent pressures of the Co and Fe effusion cells were adjusted to target the nominal Fe doping ratio x . The actual doping ratio was determined using x-ray diffraction measurements (see Sec. II in Supplemental Material [29]). The x-ray diffraction measurements were conducted after the STM measurements on the respective sample.

2. STM measurements

The as-grown $\text{Co}_{1-x}\text{Fe}_x\text{Sn}$ thin films were *in situ* transferred to a home-built STM system without breaking the ultrahigh vacuum (UHV). No postannealing of the thin films after growth was conducted. STM measurements were performed at cryogenic temperatures ($4 \text{ K} \leq T \leq 24 \text{ K}$) and UHV conditions ($p \leq 1.4 \times 10^{-10} \text{ mbar}$) using a chemically etched tungsten STM tip. The tip was prepared on a Cu(111) surface by field emission and controlled indentation and calibrated against the Cu(111) Shockley surface state before each set of measurements. dI/dV spectra were recorded using standard lock-in techniques with a small bias modulation V_m chosen between $1 \text{ mV} \leq V_m \leq 10 \text{ mV}$ at a frequency $f = 3.971 \text{ kHz}$. All dI/dV maps were recorded using the multipass mode to avoid set-point effects. To minimize piezo drift during data acquisition in multipass mode, higher current set points were chosen in order to maintain sufficient signal-to-noise ratio in the dI/dV maps while reducing the measurement time. Care was taken that the junction always remains deep inside the tunnel regime. The set-point tunnel current I , bias voltage V , lock-in modulation V_m , and temperature T of each measurement are indicated in the respective figure captions. Where applicable,

dI/dV maps were filtered for high-frequency noise stemming from mechanical vibrations coupling to the tip-sample junction of the STM.

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