

Magnetic coupled electronic landscape in bilayer-distorted titanium-based kagome metals

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Quantum materials whose atoms are arranged on a lattice of corner-sharing triangles, i.e., the kagome lattice, have recently emerged as a captivating platform for investigating exotic correlated and topological electronic phenomena. Here, we combine ultralow temperature angle-resolved photoemission spectroscopy (ARPES) with scanning tunneling microscopy and density functional theory calculations to reveal the fascinating electronic structure of the bilayer-distorted kagome material LnTi_3Bi_4 , where Ln stands for Nd and Yb. Distinct from other kagome materials, LnTi_3Bi_4 exhibits twofold, rather than sixfold, symmetries, stemming from the distorted kagome lattice, which leads to a unique electronic structure. Combining experiment and theory we map out the electronic structure and discover double flat bands as well as multiple Van Hove singularities (VHSs), with one VHS exhibiting higher-order characteristics near the Fermi level. Notably, in the magnetic version NdTi_3Bi_4 , the ultralow base temperature ARPES measurements unveil an unconventional band splitting in the band dispersions which is induced by the ferromagnetic ordering. These findings reveal the potential of bilayer-distorted kagome metals LnTi_3Bi_4 as a promising platform for exploring novel emergent phases of matter at the intersection of strong correlation and magnetism.

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In recent years, there has been a burgeoning interest in two-dimensional (2D) quantum materials, where the 2D confinement of the electrons leads to enhanced many-body effects and interesting symmetry-breaking states [1–8]. In these 2D materials a diverse set of intriguing quantum states has been observed, ranging from magnetism, charge density wave (CDW) order, and unconventional superconductivity to

electronic nematicity and beyond [1–8]. Among these 2D materials, compounds composed of atoms arranged as a lattice of corner-sharing triangles, known as the kagome lattice, have emerged as a particularly captivating platform for exploring exotic correlated and topological electronic states [2,3,6–17].

Due to the intrinsic geometric frustration, the idealized 2D kagome network possesses intriguing electronic characteristics, such as flat bands, Dirac fermions, and Van Hove singularities (VHSs). Recent experimental endeavors have primarily focused on transition-metal-based kagome magnets, exemplified by Fe_mSn_n ($m:n = 3:2, 1:1$), $\text{Co}_3\text{Sn}_2\text{S}_2$, and RMn_6Sn_6 (where R denotes a rare-earth element) [6,9–17], wherein the kagome layers are composed of magnetic atoms. Notably, in addition to the expected flat bands and Dirac fermions inherent to the hexagonal kagome structure [6,13,15], these materials have also been found to exhibit non-trivial Chern gaps [12] and relativistic Weyl fermions [9–11]. The interplay between such topological quantum states and

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intrinsic magnetism gives rise to a large anomalous Hall effect [9], underscoring the intriguing potential of these kagome magnets. Moreover, a recently discovered class of nonmagnetic kagome superconductors, such as AV_3Sb_5 (where A can be K, Rb, or Cs), hosts a diverse nature of VHSs in the vicinity of the Fermi level (E_F) [18,19]. The large density of states (DOS) near these VHSs enhances correlations, which may lead to an intricate intertwining of CDW order and electronic nematicity [18–22]. Furthermore, by replacing vanadium with a kagome network of titanium atoms, a family of kagome superconductors, ATi_3Bi_5 , was found to manifest electronic nematicity even in the absence of CDW order [23–25]. Nonmagnetic kagome lattices (whether vanadium- or titanium-based) present exciting prospects for exploring correlated quantum phenomena. Their derivatives, such as RV_6Sn_6 [26–28] and LnM_3X_4 (where Ln represents lanthanide; M involves V, Ti; X denotes Sb, Bi) [29–31], exhibit intriguing potential by introducing an anisotropic distortion to the kagome lattice and incorporating magnetic layers, thereby offering enhanced versatility and tunability and opening up intriguing avenues for exploration.

$LnTi_3Bi_4$ (where Ln can be Nd, Yb, Pr, etc.) is a recently synthesized titanium-based bilayer kagome metal [29–31], distinguished by its slightly distorted kagome lattices and zigzag Ln chains in $LnBi$ bilayers. Despite the lack of long-range magnetic order in the kagome layer, akin to its analogs ATi_3Bi_5 , the ability to introduce magnetism via controlling the Ln sites offers a flexible platform for exploring inherent kagome lattice characteristics such as correlation effects, nontrivial band topology, and VHSs in combination with magnetism. Given the unique characteristics of distorted kagome lattices and tunable magnetism in zigzag sublattices, the exploration of the electronic structure of such titanium-based kagome metal is desirable to showcase a prototypical electronic manifestation of novel Bloch eigenstates of the kagome lattice and to elucidate how magnetism manipulates the band topology, and as such to establish a paradigm in the vibrant domain of kagome quantum materials.

In this work, we combine ultralow temperature angle-resolved photoemission spectroscopy (ARPES), scanning tunneling microscopy (STM), and density functional theory (DFT) calculations to comprehensively examine the titanium-based bilayer kagome magnet $NdTi_3Bi_4$ alongside its nonmagnetic counterpart, $YbTi_3Bi_4$. Setting it apart from other kagome materials, $LnTi_3Bi_4$ exhibits a unique electronic structure characterized by twofold symmetries (C_{2x} and C_{2z}) originating from the distorted kagome lattice. We reveal the presence of double flat bands and multiple VHSs, with one VHS very close to the E_F displaying higher-order characteristics. Additionally, the ultralow base temperature of the ARPES measurement enables us to observe an unusual band splitting of the band dispersion in the kagome magnet $NdTi_3Bi_4$ below its ferromagnetic (FM) ordering temperature. These findings underscore the potential of bilayer distorted kagome metals $LnTi_3Bi_4$ as a promising platform for exploring new emergent phases of matter at the intersection of strong correlation and magnetism.

$LnTi_3Bi_4$ possesses a layered crystal structure following the orthogonal space group $Fmmm$ (no. 69). It consists of Ti_3Bi_4 kagome layers alternating with $LnBi_2$ layers along the

c axis [Figs. 1(a)(i) and 1(a)(ii)]. In contrast to the hexagonal structures of AV_3Sb_5 or ATi_3Bi_5 [Fig. 1(b)(i)], $LnTi_3Bi_4$ features bilayer kagome layers within its primitive cell. Notably, these kagome layers display an additional anisotropic distortion, resulting in a transformation of the crystal symmetry from hexagonal to orthorhombic. Consequently, in our analysis we adopt the three-dimensional orthorhombic bulk Brillouin zone (BZ) [Fig. 1(a)(iii)], while the corresponding projected 2D BZ can be considered as being a pseudo-hexagonal; see the Γ , M , K , M' and K' high-symmetry points in Figs. 1(a)(iii), 1(b)(ii), and 1(b)(iii). The reduction of crystal symmetry derived from the lattice distortion can influence the electronic structure features of the kagome lattice, such as Dirac cones and flat bands. Considering a single-orbital tight-binding model without spin-orbit coupling, a pristine single-layer kagome lattice [Fig. 1(b)(i)] is expected to host a gapless Dirac cone (protected by crystal symmetry) and a flat band, as illustrated in Fig. 1(c)(i). However, when an anisotropic in-plane distortion is introduced in the single-layer kagome lattice, the sixfold rotational symmetry (C_{6z}) is broken while the twofold rotational symmetry (C_{2z}) is preserved [Fig. 1(b)(ii)], leading to the displacement of Dirac cone away from the K and K' points, creating a gap feature at these points, and a slight dispersion in the flat band [Fig. 1(c)(ii)] [6]. In the scenario of a bilayer-distorted kagome lattice [Fig. 1(b)(iii)], the interlayer coupling between the two kagome layers further enriches the electronic structure, giving rise to double flat bands arising from bonding-antibonding splitting [Fig. 1(c)(iii)].

To explore the lattice-driven electronic confinement and double flat bands, we focus on two key members within the $LnTi_3Bi_4$ family: $NdTi_3Bi_4$, an FM kagome metal with a critical transition temperature (T_c) of 8.5 K, and the nonmagnetic $YbTi_3Bi_4$ (see Supplemental Material Fig. S1 for sample details [32]). We first investigate the electronic structure of the two compounds through DFT calculations. In line with our earlier findings in the titanium-based single-layer kagome metal ATi_3Bi_5 [25], the theoretical orbital-resolved band dispersions, for both $NdTi_3Bi_4$ [Fig. 1(d)] and $YbTi_3Bi_4$ [Fig. 1(e)], display multiple d -orbital characteristics near the E_F . However, a distinct departure from previously studied kagome compounds is apparent in the overall band dispersions of $NdTi_3Bi_4$ and $YbTi_3Bi_4$, which display a highly anisotropic nature. This anisotropy is evident when comparing two high-symmetry cuts taken along the Γ - M - K' - Γ and Γ - M' - K - Γ directions [Figs. 1(d) and 1(e)], with Γ - M and Γ - K lines serving as twofold rotational axes. Remarkably, the DFT calculations not only validate the expected presence of double flat bands below E_F , as illustrated in Fig. 1(b)(iii), but also unveil multiple VHSs in the vicinity of E_F at the M and M' points. A detailed analysis of orbital character and band parity pinpoints the $d_{xy}/d_{x^2-y^2}$ orbitals as the primary contributors to these double flat bands [Figs. 1(d)(i) and 1(e)(i)]. Furthermore, the DOS calculation reveals a notable peak within the binding energy (E_B) range 0.4–0.8 eV, further confirming the assignment of the $d_{xy}/d_{x^2-y^2}$ bands to the discussed double flat bands [highlighted by the grey stripe in Figs. 1(d) and 1(e)]. These theoretical insights align consistently with our experimental observations, as we will demonstrate in the following.

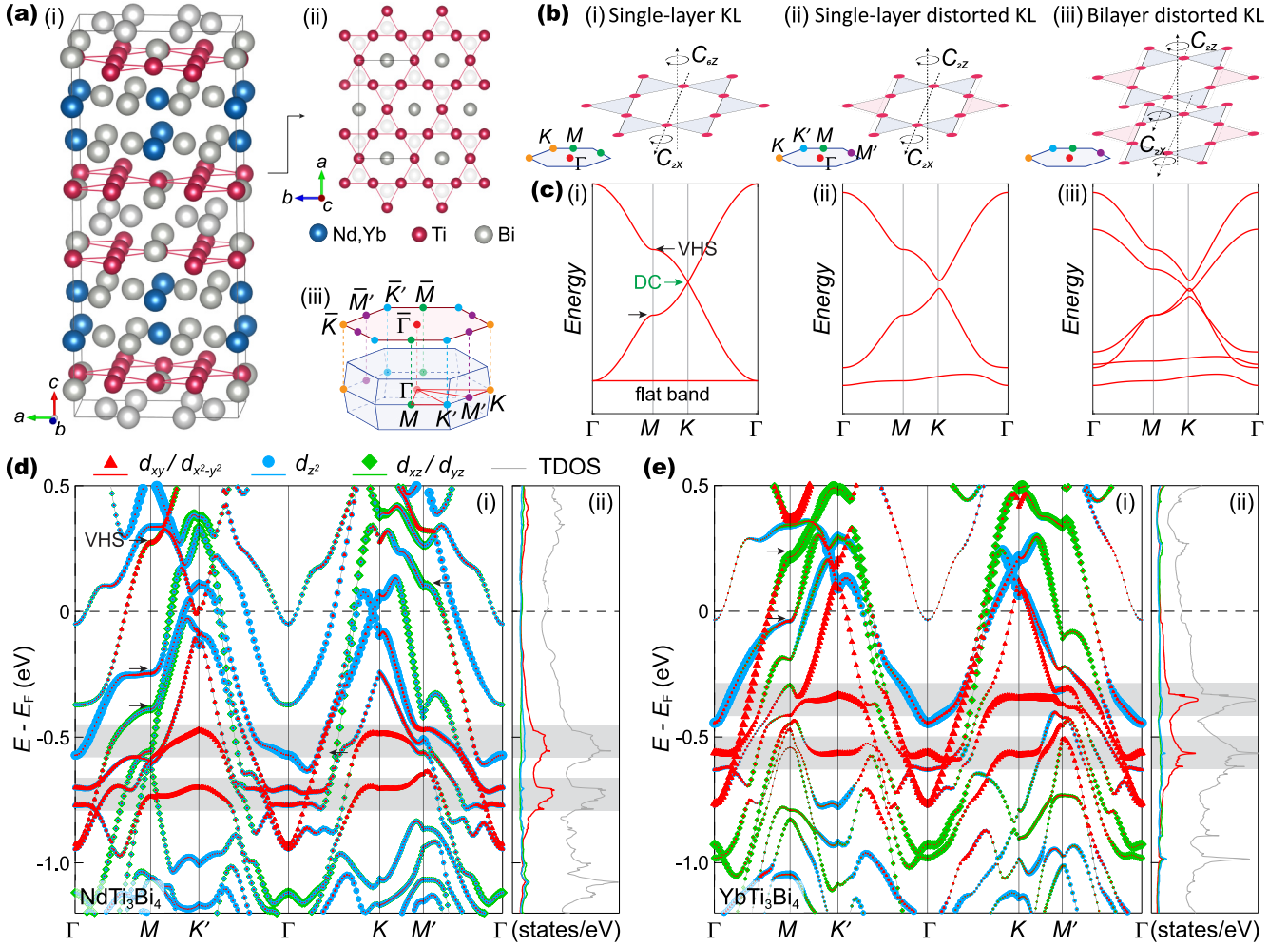


FIG. 1. Crystal structure and calculated band structure of $LnTi_3Bi_4$. (a) The unit cell of $LnTi_3Bi_4$ (i), a top view displaying the kagome plane (ii), and the bulk BZ of $LnTi_3Bi_4$ with the corresponding projected BZ (iii). (b) Twofold (C_{2x}) and sixfold (C_{6z}) rotation-axis symmetries of a pristine single kagome layer (KL) (i), twofold symmetries (C_{2x} and C_{2z}) of the single-layer-distorted KL (ii), and bilayer-distorted KL (iii). (c) A single-orbital tight-binding model without spin-orbit coupling for the single KL (i), single-layer-distorted KL (ii), and bilayer-distorted KL (iii). (d), (e) DFT calculations representing orbital character-resolved band structures of $NdTi_3Bi_4$ [(d)(i)] and $YbTi_3Bi_4$ [(e)(i)] alongside their corresponding DOS calculations [(d)(ii), (e)(ii)].

Figure 2(a) presents atomically resolved STM topography measured on the surface of $NdTi_3Bi_4$, clearly revealing a hexagonal lattice formed by the Nd atoms. We note that only the Nd-terminated layer remains robust and dominant on the cleaved sample surface. To gain insight into the electronic structure of the system, we utilize quasiparticle interference (QPI) imaging, which manifests as static modulations in $dI/dV(r, V)$ maps. The Fourier transforms of these maps show an isotropic scattering pattern, displaying an apparent mirror symmetry [Fig. 2(b) and Fig. S2]. Further exploring the isotropic nature of the electronic structure in $NdTi_3Bi_4$, Fig. 2(c)(i) showcases the experimental Fermi surface (FS) measured by ARPES on the Nd-terminated surface. It is evident that the FS topology exhibits a twofold symmetry, disregarding the spectral intensity due to its sensitivity to the matrix element effect. Remarkably, this experimental kagome fermiology aligns well with the DFT calculations depicted in Fig. 2(c)(ii). Moreover, the experimental and theoretical FSs for $YbTi_3Bi_4$ [Fig. 2(d)] show a similar twofold

symmetry observed in $NdTi_3Bi_4$. This anisotropy in the low-energy electronic structure is further corroborated by the corresponding ARPES spectra acquired along two representative high-symmetry directions: the $\bar{M}-\bar{\Gamma}-\bar{M}$ and $\bar{M}'-\bar{\Gamma}-\bar{M}'$ paths, which are related by a sixfold rotation. These consistent observations across different samples, using different techniques, strongly suggest that the observed anisotropy in the electronic structure is due to the distorted kagome lattice contained in these systems.

We then turn to the search for characteristic band features, focusing specifically on the double flat bands and VHSs. In Figs. 3(a) and 3(c), we display the polarization-dependent band dispersions of $NdTi_3Bi_4$ and $YbTi_3Bi_4$ along the $\bar{\Gamma}-\bar{K}'$ direction, respectively. The presence of the double flat bands is clearly visible [Figs. 3(a)(i) and 3(c)(i)], particularly pronounced when probed with linear-vertical polarized light [Figs. 3(a)(iii) and 3(c)(iii)]. Our calculations well reproduce these double-flat-band features [Figs. 3(b) and 3(d)]. Intriguingly, the experimental double flat bands in $NdTi_3Bi_4$

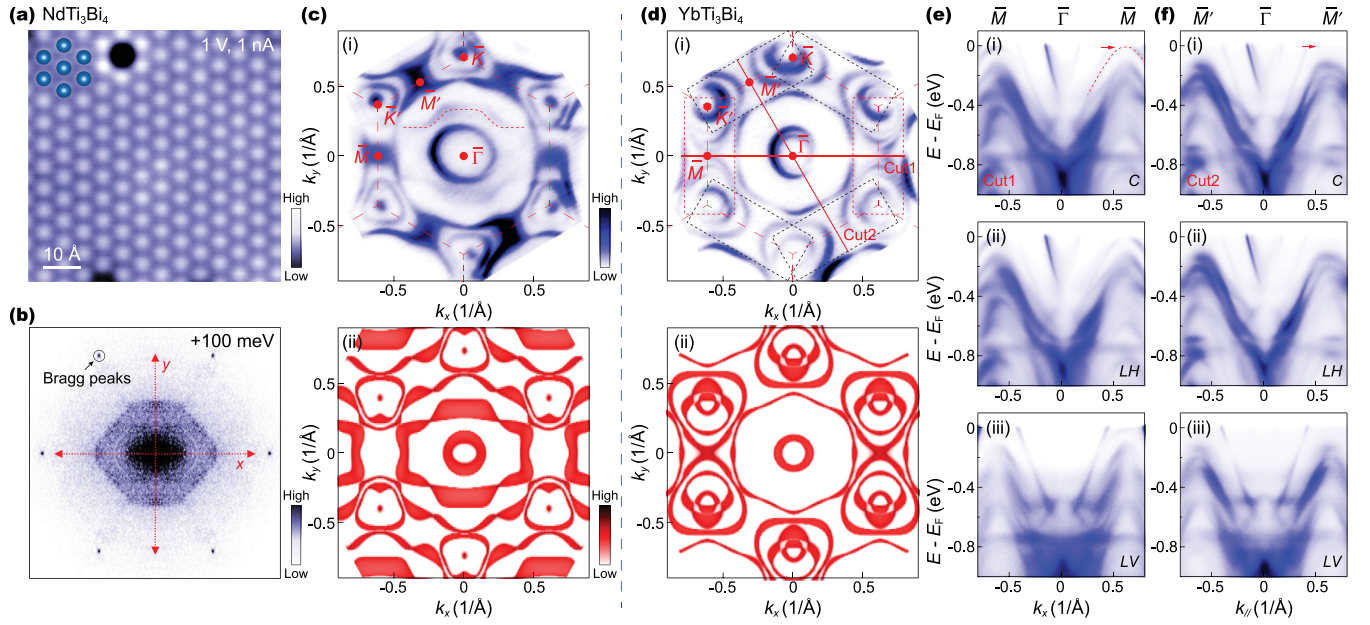


FIG. 2. Anisotropic electronic structure in LnTi_3Bi_4 . (a),(b) A STM topograph of Nd termination measured at 5 K (a) and corresponding Fourier transforms of dI/dV map at +100 meV (b). Atomic Bragg peaks are highlighted with a black ring. (c) Experimental constant energy contour (CEC) (i) and theoretical CEC (ii) of NdTi_3Bi_4 . (d) Same data as in (c), but for YbTi_3Bi_4 . (e), (f) ARPES spectra along the $\bar{\Gamma}-\bar{M}$ (e) and $\bar{\Gamma}-\bar{M}'$ (f) directions [red lines in (d)(i)], probed with circularly (C) (i), linear horizontally (LH) (ii), and linear vertically (LV) (iii) polarized lights. The k_z dispersion appears to be very weak (see Fig. S3).

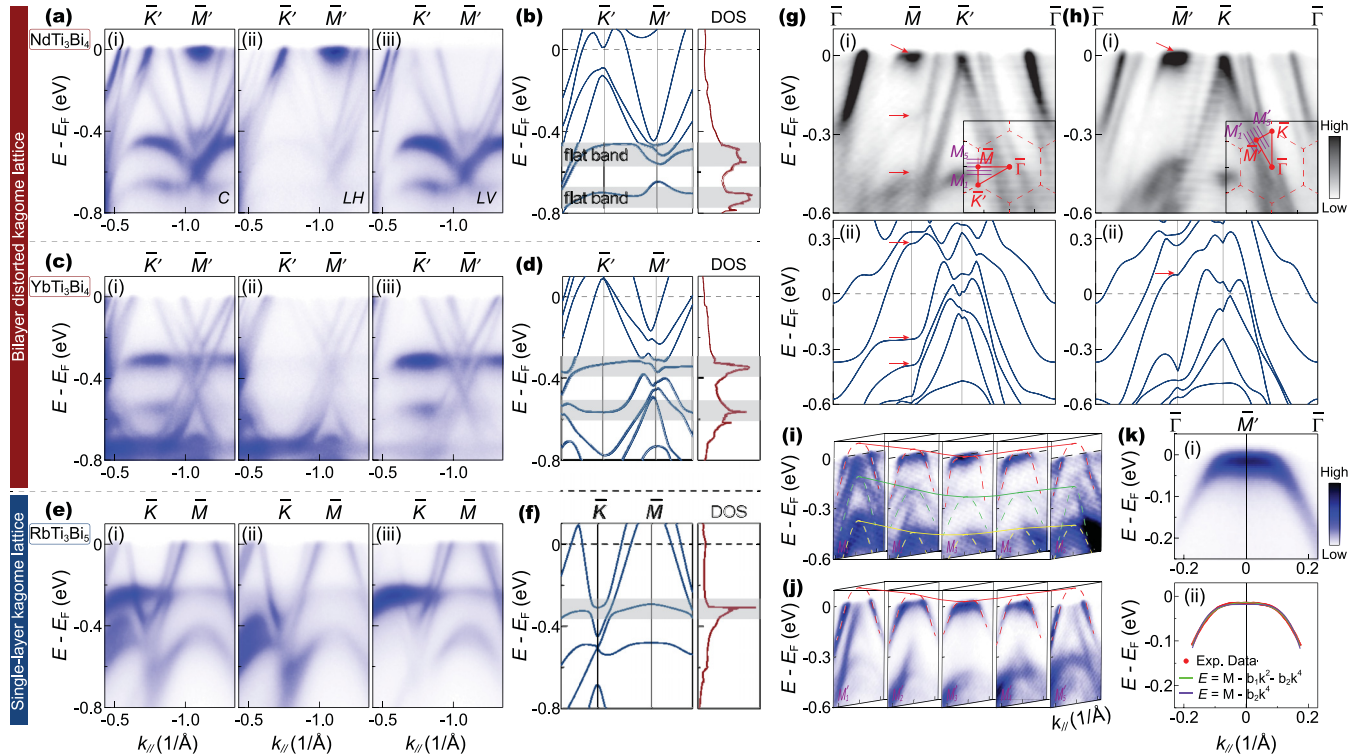


FIG. 3. Double flat bands and multiple VHSs in LnTi_3Bi_4 . (a) Experimental band dispersions of NdTi_3Bi_4 measured with C (i), LH (ii), and LV (iii) polarized lights along the $\bar{\Gamma}-\bar{K}'-\bar{M}'$ direction. The grey shading highlights the flat band. (b) DFT-calculated band structure and DOS of NdTi_3Bi_4 along the $\bar{\Gamma}-\bar{K}'-\bar{M}'$ direction. (c)-(f) Same data as in (a) and (b), but for YbTi_3Bi_4 (c,d) and RbTi_3Bi_5 (e,f). (g), (h) Experimental (i) and theoretical (ii) band dispersions of NdTi_3Bi_4 taken along the $\bar{\Gamma}-\bar{M}-\bar{K}'-\bar{\Gamma}$ (g) and $\bar{\Gamma}-\bar{M}'-\bar{K}'-\bar{\Gamma}$ (h) directions [red lines in Fig. 1(a)(iii)]. The arrows indicate the VHSs. (i), (j) ARPES spectra taken vertically across the $\bar{M}-\bar{K}'$ (i) and $\bar{M}'-\bar{K}'$ (j) paths, with the paths of the band dispersions ($\#M_1-\#M_5$) indicated by the purple lines in the insets of (g) and (h). (i) Experimental band dispersion of NdTi_3Bi_4 along the $\bar{\Gamma}-\bar{M}'$ direction (i), fittings of the measured dispersion by $E = M - b_1k^2 - b_2k^4$ and $E = M - b_2k^4$ forms (ii).

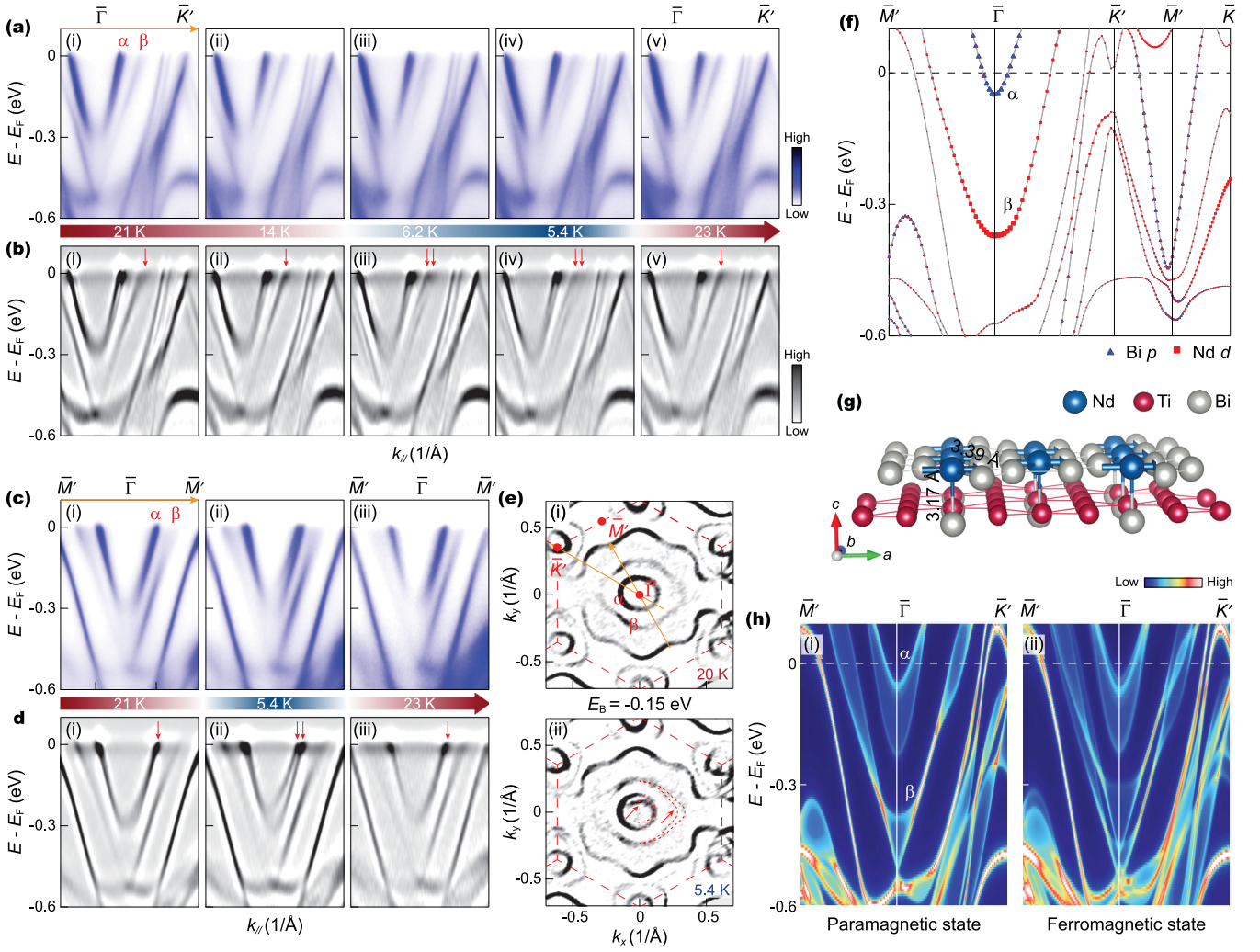


FIG. 4. Magnetism-driven orbital-selective band splittings in NdTi₃Bi₄. (a), (b) Temperature evolution of the band dispersions along the $\bar{\Gamma}$ - $\bar{K}'$ direction, probed with C polarized light (a), and their corresponding second-derivative plots with respect to energy (b). (c), (d) Same data as in (a) and (b), but measured along the $\bar{\Gamma}$ - $\bar{M}'$ direction. (e) CEC taken at $E_B = 0.15$ eV above (i) and below (ii) the magnetic transition temperature. The yellow lines mark the momentum path of the band dispersion shown in (a)–(d). The red dashed curves and arrows highlight the orbital-selective anisotropic band splittings. (f) Nd-*d* and Bi-*p* orbital-resolved DFT band structure. (g) Ferromagnetic NdBi₂ layer and its adjacent Ti₃Bi₄ kagome layer. (h) Calculated projection of bulk states in paramagnetic state (i) and FM state by introducing a FM order within the Nd *d* orbitals and the Bi *p* orbitals bonded to Nd atoms (ii).

[Fig. 3(a)] exhibit more apparent dispersions as compared to those in YbTi₃Bi₄ [Fig. 3(c)]. This disparity can be attributed to the stronger interorbital hopping embedded in the kagome sublattice of NdTi₃Bi₄. Nonetheless, the observation of double flat bands in NdTi₃Bi₄ and YbTi₃Bi₄ [Figs. 3(a)–3(d)], in contrast to the single flat band observed in the single kagome network RbTi₃Bi₅ [Figs. 3(e) and 3(f)], signifies the unique kagome-driven electronic confinement and hints at a potentially stronger electronic correlation effect in the double kagome motif if both flat bands are tuned closer to the E_F .

The correlation effect is also embodied in the VHSs derived from the kagome lattice. Figures 3(g) and 3(h) show the energy-momentum dispersions of NdTi₃Bi₄ along the $\bar{\Gamma}$ - $\bar{M}$ - $\bar{K}'$ - $\bar{\Gamma}$ [Fig. 3(g)] and $\bar{\Gamma}$ - $\bar{M}'$ - $\bar{K}$ - $\bar{\Gamma}$ [Fig. 3(h)] directions from photoemission experiment, identifying multiple VHSs at the BZ boundary ($\bar{M}$ and $\bar{M}'$). The saddle point

nature of these VHSs is further visualized through a series of cuts taken vertically across the $\bar{K}'\bar{M}$ [$\#M_1$ – $\#M_5$, as indicated in the inset of Fig. 3(g)(i)] and $\bar{K}\bar{M}'$ paths [$\#M'_1$ – $\#M'_5$ indicated in Fig. 3(h)(i)], where the band dispersions are holelike along the $\bar{M}\bar{\Gamma}$ / $\bar{M}'\bar{\Gamma}$ direction but electronlike along the $\bar{M}\bar{K}'$ / $\bar{M}'\bar{K}$ direction. The band tops of the holelike bands [dashed curves in Figs. 3(i) and 3(j)] exhibit minimum energies at the $\bar{M}$ and $\bar{M}'$ points (solid curves). The experimental VHS bands are overall consistent with the theoretical calculations [Figs. 3(g)(ii) and 3(h)(ii)], including the distinct anisotropies in band dispersions along the two high-symmetry directions [Figs. 3(g) and 3(h)]. However, we note that the energy locations of the VHSs are not accurately captured by DFT, even including the consideration of an overall renormalization. Additionally, one VHS in the experiment, located very close to E_F , appears to exhibit a much flatter dispersion than within our DFT calculations [Fig. 3(k)(i)]. Upon

fitting the experimental band dispersion, we find that the quadratic term is much smaller than the quartic one along the $\bar{M}\bar{\Gamma}$ direction, indicating a higher-order nature of the VHS [Fig. 3(k)(ii)] [18,19]. These findings suggest that an orbital-dependent correlation effect is at play in the kagome magnet NdTi_3Bi_4 .

Finally, we investigate how magnetism modifies the electronic structure in NdTi_3Bi_4 , through detailed temperature-dependent ARPES measurements spanning across the magnetic transition temperature T_c . As summarized in Figs. 4(a) and 4(b), upon cooling, the band dispersions initially display minimal changes [Figs. 4(a)(i), 4(a)(ii), 4(b)(i), and 4(b)(ii)]. As the temperature continues to decrease, however, a noticeable splitting in the β band along the $\bar{\Gamma}\text{--}\bar{K}'$ direction becomes evident [Figs. 4(a)(iii), 4(a)(iv)(iv), 4(b)(iii), and 4(b)(iv)]. Upon warming, this splitting vanishes [Figs. 4(a)(v) and 4(b)(v)], and the bands revert to their initial states [Figs. 4(a)(i) and 4(b)(i)] and Fig. S4]. A similar band splitting also occurs in the temperature evolution of the band structure along the $\bar{\Gamma}\text{--}\bar{M}'$ direction [Figs. 4(c) and 4(d)]. Intriguingly, the band splitting manifests in a different band, namely, the α band. Furthermore, a side-by-side comparison between the constant energy contour take at $E_B = 0.15$ eV above and below T_c clearly reveals highly anisotropic band splittings in the magnetic state [Fig. 4(e)]. Specifically, the α band exhibits maximum splitting along the $\bar{\Gamma}\text{--}\bar{M}$ direction and minimal splitting along the $\bar{\Gamma}\text{--}\bar{K}$ direction, whereas the β band displays the largest splitting along the $\bar{\Gamma}\text{--}\bar{K}$ direction and less pronounced splitting along the $\bar{\Gamma}\text{--}\bar{M}$ direction. These contrasting band splittings imply distinct magnetic couplings within these two bands.

To understand the origin of the observed band splittings in the specific α and β bands at low temperature, we turn to theoretical modeling, starting with an exploration of the orbital-resolved band structure in the paramagnetic phase. As depicted in Fig. 4(f), the β and α bands are found to be dominantly contributed by the Nd d orbitals and the p orbitals of Bi atoms bonded to Nd atoms, respectively (for details, see Fig. S5). These orbital characteristics, coupled with the coincidence of the splitting temperature and T_c , near 8.5 K, suggest that the observed orbital-selective splittings are intimately related to the formation of FM order in the NdBi_2 layer [Fig. 4(g)]. To simulate the effects of FM order, we calculate the band structures of NdTi_3Bi_4 by including a FM splitting within the Nd d orbitals and the Bi p orbitals that are bonded to the Nd atoms. Remarkably, these calculations replicate well the orbital-selective splittings [Fig. 4(h)] even though the theoretical calculations do not entirely capture the pronounced anisotropic nature of the splittings. One possible explanation for this discrepancy is the enhanced impact of the

spin-orbit coupling associated with the surface NdBi_2 layers, a factor that has not been taken into account in our calculations.

In summary, we have investigated the intricate electronic band structure of the distorted bilayer kagome material LnTi_3Bi_4 , which is characterized by twofold symmetries due to the lattice distortion. In particular, we have unveiled the presence of double flat bands and multiple VHSs, one of which exhibits a higher-order nature in close proximity to E_F . Additionally, we have observed an unconventional band splitting in the band dispersions of the kagome magnet NdTi_3Bi_4 below its FM transition temperature. The discovery of the double kagome flat bands calls for further research to attempt raising them to the E_F , which would allow us to look for the proposed lattice-borne correlated topological phases [33]. While the origin of the anisotropic band splitting warrants further investigation, our results point toward the potential for generating magnetism-driven emergent spin-splitting surface states in the context of kagome materials [34]. Collectively, our findings establish the bilayer-distorted kagome metals LnTi_3Bi_4 as a promising platform for studying the intricate interplay between lattice distortion, electronic correlations, and magnetism within the framework of a distorted kagome lattice.

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Y.H. conceived the ARPES experiments. L.C. grew and characterized the crystals with guidance from Y.Z. and G.W. C.L. and X.W. performed the theoretical calculations with support from R.T. and A.P.S. Y.H. performed the ARPES experiments with support from N.C.P., M.R., and M.S. H.D. and J.-X.Y. performed the STM experiments. M.R. and N.C.P. maintained the ARPES facilities at the SIS-ULTRA. Y.H. analyzed the data. Y.H. wrote the manuscript with inputs from all authors. M.S., X.W., and Y.H. supervised the project.

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- [1] B. Keimer and J. Moore, The physics of quantum materials, *Nat. Phys.* **13**, 1045 (2017).
 - [2] M. L. Kiesel and R. Thomale, Sublattice interference in the kagome Hubbard model, *Phys. Rev. B* **86**, 121105(R) (2012).
 - [3] M. L. Kiesel, C. Platt, and R. Thomale, Unconventional Fermi surface instabilities in the kagome Hubbard model, *Phys. Rev. Lett.* **110**, 126405 (2013).
 - [4] W.-S. Wang, Z.-Z. Li, Y.-Y. Xiang, and Q.-H. Wang, Competing electronic orders on kagome lattices at van Hove filling, *Phys. Rev. B* **87**, 115135 (2013).
 - [5] Y. Cao, V. Fatemi, S. Fang, K. Watanabe, T. Taniguchi, E. Kaxiras, and P. Jarillo-Herrero, Unconventional superconductivity in magic-angle graphene superlattices, *Nature (London)* **556**, 43 (2018).

- [6] L. Ye, M. Kang, J. Liu, F. Cube, C. R. Wicker, T. Suzuki, C. Jozwiak, A. Bostwick, E. Rotenberg, D. C. Bell *et al.*, Massive Dirac fermions in a ferromagnetic kagome metal, *Nature (London)* **555**, 638 (2018).
- [7] B. R. Ortiz, S. M. L. Teicher, Y. Hu, J. L. Zuo, P. M. Sarte, E. C. Schueller, A. M. Milinda Abeykoon, M. J. Krogstad, S. Rosenkranz, R. Osborn, R. Seshadri, L. Balents, J. He, and S. D. Wilson, CsV₃Sb₅: A $\mathbb{Z}_2$ Topological kagome metal with a superconducting ground state, *Phys. Rev. Lett.* **125**, 247002 (2020).
- [8] L. Nie, K. Sun, W. Ma, D. Song, L. Zheng, Z. Liang, P. Wu, F. Yu, J. Li, M. Shan, D. Zhao, S. Li, B. Kang, Z. Wu, Y. Zhou, K. Liu, Z. Xiang, J. Ying, Z. Wang, T. Wu, and X. Chen, Charge-density-wave-driven electronic nematicity in a kagome superconductor, *Nature (London)* **604**, 59 (2022).
- [9] E. Liu, Y. Sun, N. Kumar, L. Muechler, A. Sun, L. Jiao, S.-Y. Yang, D. Liu, A. Liang, Q. Xu *et al.*, Giant anomalous Hall effect in a ferromagnetic kagome-lattice semimetal, *Nat. Phys.* **14**, 1125 (2018).
- [10] N. Morali, R. Batabyal, P. K. Nag, E. Liu, Q. Xu, Y. Sun, B. Yan, C. Felser, N. Avraham, and H. Beidenkopf, Fermi-arc diversity on surface terminations of the magnetic Weyl semimetal Co₃Sn₂S₂, *Science* **365**, 1286 (2019).
- [11] D.-F. Liu, A. J. Liang, E. K. Liu, Q. N. Xu, Y. W. Li, C. Chen, D. Pei, W. J. Shi, S. K. Mo, P. Dudin, T. Kim, C. Cacho, G. Li, Y. Sun, L. X. Yang *et al.*, Magnetic Weyl semimetal phase in a kagomé crystal, *Science* **365**, 1282 (2019).
- [12] J.-X. Yin, W. Ma, T. A. Cochran, X. Xu, S. S. Zhang, H.-J. Tien, N. Shumiya, G. Cheng, K. Jiang, B. Lian *et al.*, Quantum-limit Chern topological magnetism in TbMn₆Sn₆, *Nature (London)* **583**, 533 (2020).
- [13] M. Kang, L. Ye, S. Fang, J.-S. You, A. Levitan, M. Han, J. I. Facio, C. Jozwiak, A. Bostwick, E. Rotenberg *et al.*, Dirac fermions and flat bands in the ideal kagome metal FeSn, *Nat. Mater.* **19**, 163 (2020).
- [14] N. J. Ghimire, R. L. Dally, L. Poudel, D. C. Jones, D. Michel, N. T. Magar, M. Bleuel, M. A. McGuire, J. S. Jiang, J. F. Mitchell *et al.*, Competing magnetic phases and fluctuation-driven scalar spin chirality in the kagome metal YMn₆Sn₆, *Sci. Adv.* **6**, 51 (2020).
- [15] M. Li, Q. Wang, G. Wang, Z. Yuan, W. Song, R. Lou, Z. Liu, Y. Huang, Z. Liu, H. Lei *et al.*, Spin-polarized Dirac cone, flat band and saddle point in kagome magnet YMn₆Sn₆, *Nat. Commun.* **12**, 3129 (2021).
- [16] W. Ma, X. Xu, J.-X. Yin, H. Yang, H. Zhou, Z.-J. Cheng, Y. Huang, Z. Qu, F. Wang, M. Z. Hasan, and S. Jia, Rare earth engineering in RMn₆Sn₆ ($R = \text{Gd} - \text{Tm}, \text{Lu}$) topological kagome magnets, *Phys. Rev. Lett.* **126**, 246602 (2021).
- [17] Y. Wang, H. Wu, G. T. McCandless, J. Y. Chan, and M. N. Ali, Quantum states and intertwining phases in kagome materials, *Nat. Rev. Phys.* **5**, 635 (2023).
- [18] Y. Hu, X. Wu, B. R. Ortiz, S. Ju, X. Han, J. Z. Ma, N. C. Plumb, M. Radovic, R. Thomale, S. D. Wilson *et al.*, Rich nature of van Hove singularities in kagome superconductor CsV₃Sb₅, *Nat. Commun.* **13**, 2220 (2022).
- [19] M. Kang, S. Fang, J. Kim, B. R. Ortiz, S. H. Ryu, J. Kim, J. Yoo, G. Sangiovanni, D. Di Sante, B.-G. Park *et al.*, Twofold van Hove singularity and origin of charge order in topological kagome superconductor CsV₃Sb₅, *Nat. Phys.* **18**, 301 (2022).
- [20] X. Wu, T. Schwemmer, T. Müller, A. Consiglio, G. Sangiovanni, D. Di Sante, Y. Iqbal, W. Hanke, A. P. Schnyder, M. M. Denner *et al.*, Nature of unconventional pairing in the kagome superconductors AV₃Sb₅ ($A = \text{K}, \text{Rb}, \text{Cs}$), *Phys. Rev. Lett.* **127**, 177001 (2021).
- [21] Y. Hu, X. Wu, A. P. Schnyder, and M. Shi, Electronic landscape of kagome superconductors AV₃Sb₅ ($A = \text{K}, \text{Rb}, \text{Cs}$) from angle-resolved photoemission spectroscopy, *npj Quantum Mater.* **8**, 67 (2023).
- [22] J.-X. Yin, B. Lian, and M. Z. Hasan, Topological kagome magnets and superconductors, *Nature (London)* **612**, 647 (2022).
- [23] H. Yang, Y. Ye, Z. Zhao, J. Liu, X.-W. Yi, Y. Zhang, J. Shi, J.-Y. You, Z. Huang, B. Wang *et al.*, Superconductivity and orbital-selective nematic order in a new titanium-based kagome metal CsTi₃Bi₅, *arXiv:2211.12264*.
- [24] H. Li, S. Cheng, B. R. Ortiz, H. Tan, D. Werhahn, K. Zeng, D. Johrendet, B. Yan, Z. Wang, S. D. Wilson *et al.*, Electronic nematicity in the absence of charge density waves in a new titanium-based kagome metal, *Nat. Phys.* **19**, 1591 (2023).
- [25] Y. Hu, C. Le, Y. Zhang, Z. Zhao, J. Liu, Z. Ma, N. C. Plumb, M. Radovic, H. Chen, A. P. Schnyder *et al.*, Non-trivial band topology and orbital-selective electronic nematicity in a titanium-based kagome superconductor, *Nat. Phys.* **19**, 1827 (2023).
- [26] G. Pokharel, S. M. L. Teicher, B. R. Ortiz, P. M. Sarte, G. Wu, S. Peng, J. He, R. Seshadri, and S. D. Wilson, Electronic properties of the topological kagome metals YV₆Sn₆ and GdV₆Sn₆, *Phys. Rev. B* **104**, 235139 (2021).
- [27] H. W. S. Arachchige, S. Arachchige, W. R. Meier, M. Marshall, T. Matsuoka, R. Xue, M. A. McGuire, R. P. Hermann, H. Cao, and D. Mandrus, Charge density wave in kagome lattice intermetallic ScV₆Sn₆, *Phys. Rev. Lett.* **129**, 216402 (2022).
- [28] Y. Hu, X. Wu, Y. Yang, S. Gao, N. C. Plumb, A. P. Schnyder, W. Xie, J. Ma, and M. Shi, Tunable topological Dirac surface states and van Hove singularities in kagome metal GdV₆Sn₆, *Sci. Adv.* **8**, add2024 (2022).
- [29] L. Chen, Y. Zhou, H. Zhang, X. Ji, K. Liao, Y. Ji, Y. Li, Z. Guo, X. Shen, R. Yu *et al.*, Tunable magnetism in Titanium-based kagome metals by rare-earth engineering and high pressure, *Commun. Mater.* **5**, 73 (2024).
- [30] Z. Zheng, L. Chen, X. Ji, Y. Zhou, G. Qu, M. Hu, Y. Huang, H. Weng, T. Qian, and G. Wang, Anisotropic magnetism and band evolution induced by ferromagnetic phase transition in titanium-based kagome ferromagnet SmTi₃Bi₄, *Sci. China Phys. Mech. Astron.* **67**, 267411 (2024).
- [31] B. R. Ortiz, H. Miao, D. S. Parker, F. Yang, G. D. Samolyuk, E. M. Clements, A. Rajapitamahuni, T. Yilmaz, E. Vescovo, J. Yan *et al.*, Evolution of highly anisotropic magnetism in the titanium-based kagome metals LnTi₃Bi₄ ($\text{Ln}: \text{La} \dots \text{Gd}^{3+}, \text{Eu}^{2+}, \text{Yb}^{2+}$), *Chem. Mater.* **35**, 9756 (2023).
- [32] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/PhysRevB.110.L121114> for methods, sample characterizations, anisotropic electronic structure revealed by STM, and other supporting information, which includes Refs. [35–40].
- [33] N. Regnault, Y. Xu, M.-R. Li, D.-S. Ma, M. Jovanovic, A. Yazdani, S. S. P. Parkin, C. Felser, L. M. Schoop, N. P. Ong *et al.*, Catalogue of flat-band stoichiometric materials, *Nature (London)* **603**, 824 (2022).

- [34] A. Chikina, M. Höppner, S. Seiro, K. Kummer, S. Danzenbächer, S. Patil, A. Generalov, M. Güttler, Y. Kucherenko, and E. V. Chulkov, Strong ferromagnetism at the surface of an antiferromagnet caused by buried magnetic moments, *Nat. Commun.* **5**, 3171 (2014).
- [35] G. Kresse and J. Hafner, *Ab initio* molecular dynamics for liquid metals, *Phys. Rev. B* **47**, 558 (1993).
- [36] G. Kresse and J. Furthmüller, Efficiency of *ab-initio* total energy calculations for metals and semiconductors using a plane-wave basis set, *Comput. Mater. Sci.* **6**, 15 (1996).
- [37] G. Kresse and J. Furthmüller, Efficient iterative schemes for *ab initio* total-energy calculations using a plane-wave basis set, *Phys. Rev. B* **54**, 11169 (1996).
- [38] H. J. Monkhorst and J. D. Pack, Special points for Brillouin-zone integrations, *Phys. Rev. B* **13**, 5188 (1976).
- [39] A. A. Mostofi, J. R. Yates, Y.-S. Lee, I. Souza, D. Vanderbilt, and N. Marzari, wannier90: A tool for obtaining maximally-localised Wannier functions, *Comput. Phys. Commun.* **178**, 685 (2008).
- [40] M. P. Lopez Sancho, J. M. Lopez Sancho, J. M. L. Sancho, and J. Rubio, Highly convergent schemes for the calculation of bulk and surface Green functions, *J. Phys. F: Met. Phys.* **15**, 851 (1985).