S1. p-type and m-type VHSs in GdV₆Sn₆

The vanadium kagome lattice of the kagome metal GdV₆Sn₆ hosts three distinct sublattices located at 3f Wyckoff positions, as illustrated in Fig. S1a. The toy band of the kagome lattice features two different types of van Hove singularities (VHSs): p-type and m-type. For the p-type VHS, the eigenstates near the three M points are contributed by mutually different sublattices [Fig. S1b(i)], while the states of the m-type VHS are equally distributed over mutually different sets of two sublattices for each M point [Fig. S1b(ii)]. The orbitally decomposed electronic structure of GdV₆Sn₆ from DFT calculations is displayed in Fig. S1c, where four VHS points occur at the M point in the vicinity of the Fermi level (E_F) (indicated by the red arrows and labeled as VHS1-4). The states of VHS1, VHS2 and VHS3 at the M point are characterized by V dₓz, dᵧy and dₓz orbitals, and are solely attributed to one sublattice in the V kagome lattice. Thus, the three VHSs are of p-type, while the states of VHS4 are attributed to a mixture of two sublattices and VHS4 is of m-type.

Fig. S1|Sublattice feature and VHSs in the kagome metals GdV₆Sn₆. a Real space structure of the vanadium kagome planes. The red, blue and green coloring indicates the three kagome sublattices. b Two distinct types of sublattice decorated van Hove singularities (VHSs), labeled as p-type [sublattice pure, (i)] and m-type [sublattice mixing, (ii)]. c DFT calculations for the orbital character resolved band structure of GdV₆Sn₆.
S2. Density functional theory calculations for surface states in GdV₆Sn₆

We have directly studied the surface states on (001) surface of GdV₆Sn₆ by performing calculations on a slab with two kind of surface terminations, which contains 10 V kagome layers and 5 GdSn₂ layers. As shown in Fig. S2, the electronic structure of GdV₆Sn₆ exhibits distinct surface states on the Gd (the red curves in Figs. S2a and S2b) and kagome terminations (the blue curves in Figs. S2a and S2c). More interestingly, the Dirac surface states on the GdSn₂ layer bridge the large local band gap at the Γ, which represent the topologically non-trivial Dirac surface states (TDSSs) originating from a $\mathbb{Z}_2$ bulk topology. On the other hand, the in-gap surface states of the kagome termination (Fig. S2c) are much more complicated than those of Gd termination (Fig. S2b), which may be attributed to more dangling bonds on the kagome termination.

Fig. S2|Surface states decomposed band structure of GdV₆Sn₆ from DFT calculations. a Band structure of GdV₆Sn₆, with bulk states (BS) and surface states (SS) decomposed. b,c Electronic structure on GdSn₂ layer (b) and V kagome layer (c). Red and blue curves indicate the SS on GdSn₂ layer and kagome layer, respectively.
S3. Analysis of the two terminations based on XPS spectra

By using a small beam spot, we have resolved two sets of representative photoemission spectra (Fig. 2 in the main text) associated with the two types of terminations on the cleaved sample surface of GdV₆Sn₆ (as illustrated in Figs. S3a and S3e). As shown in Figs. S3b and S3f, the X-ray photoelectron spectroscopy (XPS) spectra clearly show the characteristic 3s, 3p peaks of V, 4f peak of Gd, and 4d peaks of Sn, exhibiting two distinct spectra on the Sn 4d core level (Figs. S3c and S3g). Following the strategy in the kagome lattice FeSn [7], the XPS spectra of the Sn-4d peaks are used to determine the surface terminations of the sample. It has been found in FeSn that Sn atoms in different local environments can contribute 4d peaks with different binding energies [7]. In GdV₆Sn₆, since Sn atoms near the kagome termination (Fig. S3a) have a more complex local environment than Sn atoms in the Gd termination (Fig. S3e), the XPS spectra exhibiting side peaks of Sn-4d (Figs. S3b and S3c) should be associated with the kagome termination (Fig. S3a). Therefore, we assign the XPS spectra showing more Sn-4d peaks to the V-kagome termination (Figs. S3a-d), while another set of spectra represents the Gd termination (Figs. S3e-h).

Furthermore, the intensity of the Gd-4f peak in the determined Gd termination (Fig. S3h) is stronger than that in the kagome termination (Fig. S3d). Moreover, the observed TDSSs on the determined Gd termination are in excellent agreement with the DFT calculations for GdSn₂ layer (see Fig. 3 and Fig. 4 in the main text), further confirming our assignment of termination. While all results show good agreement, the unambiguous determination of the surface termination should come from a combination of in situ scanning tunneling microscopy (STM) and ARPES techniques, which is not very necessary for the present work. The termination assignments do not affect the main conclusions of the tunable TDSSs and VHSs in GdV₆Sn₆.
Fig. S3 | Determination of surface terminations of GdV₆Sn₆ using XPS.  

a) Schematics of the kagome termination in GdV₆Sn₆. 

b) Wide-energy-range XPS spectrum of GdV₆Sn₆ measured with 170 eV photon. 

c, d) Zoom-in plot of the spectrum highlighting Sn-4d peaks (c) and Gd-4f peak (d) in selected energy regions in (b), as indicated by the black and blue box, respectively. 

e-h) Same as (a-d), but for the Gd termination.
S4. Dirac cone and VHS endemic to the kagome lattice of GdV₆Sn₆

The kagome structural motif could naturally give rise to kagome-derived electronic bands with Dirac cone and VHS. Indeed, we have identified the characteristic Dirac cone and VHS point of the kagome lattice on the kagome termination of GdV₆Sn₆ (Fig. S4). As shown in Fig. S4a, the Dirac cone is clearly observed around \( \bar{K} \) point, at the binding energy \( (E_B) \) around 200 meV (see the red arrow). Interestingly, after surface potassium deposition, the Dirac cone persists while trivial surface states [29] are killed (Fig. S4b), indicating the bulk origin and robustness of the Dirac fermions. Moreover, VHS is also revealed near \( \bar{M} \) point (Fig. S4c), as confirmed by the calculations (Fig. S4d). The identified Dirac cone and VHS on the kagome termination are consistently observed on the Gd termination, unambiguously confirming that they are bulk states native to the kagome lattice of GdV₆Sn₆ (for details, see Fig. S5).

**Fig. S4|Dirac cone and VHS in GdV₆Sn₆.** a ARPES spectra taken along the \( \bar{K} - \bar{M} - \bar{K} \) direction, on the pristine kagome surface, measured with 76 eV linear horizontal \( (LH) \) polarization. b Same as (a), but collected on the K dosed kagome surface, measured with 76 eV \( LH \) (i) and linear vertical \( (LV) \) (ii) polarizations. Red arrow highlights the Dirac cone. Arrow indicates the VHS1 band. c ARPES spectrum along the \( \bar{K} - \bar{M} - \bar{\Gamma} \) path showing the VHS1 at the \( \bar{M} \) point, obtained on kagome surface. d Same as (c), but from DFT calculations.
S5. Detailed comparison between the experimental and theoretical band dispersions

We compare the ARPES spectra obtained on the Gd termination with the DFT calculated energy band dispersions in Fig. S5. Figures S5a and S5b display the band structure of GdV₆Sn₆ in the paramagnetic phase from DFT calculations. We next focus on two high symmetry cuts along the $\Gamma - K - M - \bar{K}$ direction (the momentum path is represented by the blue line in Fig. S5c, as marked as Cut#1) and the $\Gamma - \bar{M}$ direction (Cut#2 in Fig. S5c), as shown in Figs. S5d,5e and Figs. S5f,5g, respectively. The experimental and calculated bands show good overall agreement, although there are discrepancies in Fermi levels between the experimental (the red dashed line in Figs. S5e, S5g) and calculated (the black dashed line in Figs. S5e, S5g) results. The agreement between experiments and calculations allows us to determine that the identified VHS (Fig. 2K in the main text, and Figs. S5h,5i) is corresponding to the VHS1 (Figs. S5a and S5j).

The experimentally determined orbital character of the bands that form the VHS1 is also consistent with the DFT calculations. Under our ARPES geometry (for details, see Ref. 20), according to the selection rules, the bands with $d_{xz}$ character along the $\Gamma - K$ and $\Gamma - \bar{M}$ paths are favored under the linear vertical ($LV$) and linear horizontal ($LH$) polarizations, respectively. Indeed, the VHS1 bands are observable under the $LV$ polarization in Fig. S5d(iii) and the $LH$ polarization in Fig. S5f(i). Therefore, the experimental VHS1 bands are contributed by Vanadium $d_{xz}$ orbital (as indicated in Figs. S5h,5i) and derived from kagome bulk states, which are in line with the DFT calculations for the orbital character resolved band structure (Figs. S5b and S5j). On the other hand, the determination of the orbital character (i.e., $d_{xz}$) of the VHS1 by polarization-dependent measurements confirms its $p$ type nature.
Fig. S5|Comparison between the experimental and theoretical dispersions in GdV$_6$Sn$_6$. a DFT calculated electronic structure, with VHSs indicated. b Orbitally decomposed electronic structure. c Brillouin zone (BZ) of GdV$_6$Sn$_6$, with high symmetry points marked. d ARPES spectra taken along the $\Gamma$ - $\bar{K}$ - $\bar{M}$ direction, on the pristine surface, measured with 76 eV circular (C) (i), LH (ii), LV (iii) polarizations. The momentum path for (d) is indicated by the blue line in (c). e Theoretical dispersion along the $\Gamma$ - $\bar{K}$ - $\bar{M}$ direction. f ARPES spectra taken along the $\Gamma$ - $M$ direction, measured with 76 eV LH (i), LV (ii) polarizations. g Theoretical bands along the $\Gamma$ - $M$ direction. h-j Band dispersion from the Gd termination (h), kagome termination (i), and DFT calculations (j), along the $\bar{K}$ - $\bar{M}$ - $\bar{\Gamma}$ path showing the VHS1 at the $M$ point. We note that the dispersions in (h) and (i) are slightly different, which may be due to the matrix element effects.
S6. Photon energy-dependent measurements on the surface states

Fig. S6 | Photon energy-dependent measurements on the Gd termination. a Photon energy-dependent ARPES spectra along the $\tilde{M} - \tilde{\Gamma} - \tilde{M}$ direction. Red dashed curves and arrows highlight the V-shaped TDSSs. In contrast to the bulk states, the V-shaped bands around $\Gamma$ do not disperse with photon energies, reflecting their surface nature. b ARPES spectral intensity as a function of photon energy along the $\Gamma - \Lambda$ direction. c Photon energy-dependent ARPES spectral intensity map at $E_F$ along the $M - \tilde{\Gamma} - M$ direction, where the red dashed lines indicate the TDSSs that show no dispersion along the energy (and thus $k_z$) direction. Red and yellow arrows mark the momentum direction of the spectrum in (b) and [Fig. S8(c)] respectively. d Same as (c), but measured along the $\tilde{K} - M - \tilde{K}$ direction. In contrast to the TDSSs in (c), the bulk states exhibit pronounced $k_z$ dispersions.
S7. **Projections of surface states on Gd and kagome terminated surface**

Figure S7 shows the spectra of bulk, GdSn$_2$ terminated surface and V terminated surface calculated from the Wannier functions. In the Gd termination, the topological Dirac surface states can be clearly identified (Fig. S7b) and are in good agreement with the DFT calculations (Fig. S2b). We note that the surface states on the kagome termination (Fig. S7c) are different from those in the DFT calculations (Fig. S2c), since the DFT calculations include charge self-consistency on the surface but that is not included in the tight-binding-model based calculations.

In realistic materials, the energy position of the Dirac point and shape of the TSSs can be greatly affected by the surface situations, such as surface potential, surface relaxation and reconstruction, band bending effects, etc. These may significantly modify the surface states of kagome termination, resulting quite different surface states (compared with DFT calculations). As the Gd termination may be relatively stable and such modification is quite small, the measured surface states (Fig. 3A and Fig. 4C in the main text) are in good agreement with the calculations (Fig. S7b), while significant surface relaxation or reconstruction may occur due to the dangling bonds on the kagome termination. This may render most of the surface states on kagome termination well above the $E_F$, which are beyond ARPES measurements.

![Fig. S7(001) surface states of GdV$_6$Sn$_6$ from Wannier tight-binding model. a-c](image)

**Fig. S7(001) surface states of GdV$_6$Sn$_6$ from Wannier tight-binding model. a-c** The (001) surface Green’s function projection of pure bulk states (BS)(a), the states [BS and surface states (SS)] on Gd termination (b), and the states (BS+SS) on Vanadium kagome termination (c).
S8. Spin texture of the TDSSs

We examined spin textures of the TDSSs on the GdSn$_2$ surface by spin-resolved ARPES and the DFT calculations. Spin-ARPES momentum distribution curve (MDC) measured at $T = 30\, K$ (i.e., paramagnetic state) displays clear spin-momentum locking of the V-shaped bands with polarization inversion of the $y$ component of the spin (Figs. S8a and b). The spin-ARPES determined spin polarization is consistent with the DFT calculations (Fig. S8c), suggesting that the circular-shaped and hexagonal-shaped Fermi surfaces have clockwise spin texture chirality [Fig. S8c, sketch in Fig. 3F and Fig. 4D(i) in the main text]. These results provide clear spectroscopic evidence of the non-trivial topological character of the V-shaped bands.

On the pristine GdSn$_2$ surface, only the lower branch of the TDSSs contributes to the Fermi surfaces, i.e., the circular-shaped and hexagonal-shaped pockets around $\overline{\Gamma}$ point (Fig. S8c, and Figs. 3E,3F in the main text). Upon sufficient electron doping [Fig. 4C(iii) in the main text, and Fig. S9], the Dirac point of the TDSSs can be tuned below the $E_F$. Therefore, both the lower and upper branches of the doped TDSSs contribute to the Fermi surfaces. However, the lower branch merges into the bulk bands [Fig. 4D(iii) in the main text, Figs. S8d and S9d], so that the corresponding Fermi surface has counterclockwise spin texture chirality (Fig. S8d). These results demonstrate the spin polarization reversal on the Fermi surfaces of GdV$_6$Sn$_6$ with doping.

**Fig. S8|Spin texture of the TDSSs.** a ARPES spectrum taken along the $\overline{\Gamma}$ - $\overline{M}$ direction, measured with 76 eV, circular polarized light. b Spin-resolved MDC, collected along the orange line in (a). The red and blue symbols in (i) are the intensity of the spin-up and spin-down states, respectively. The black curve indicates the spin polarization (ii). c,d Spin textures of the TDSSs on the pristine (a) and electron doped (b) GdSn$_2$ surface. Green arrow indicates the spin-polarization direction of the TDSSs.
S9. Momentum-dependent dispersions of the TDSSs

Revealing the details of the TDSSs in GdV₆Sn₆, Figure S8 displays the momentum dependence and photon energy dependence of the doped TDSSs on the K-dosed GdSn₂ surface. Figure S8a plots the constant energy contours and their evolution with binding energy ($E_B$). With increasing $E_B$, the TDSSs contributed hexagonal-shaped pockets near the zone center (Γ point) shrink (Fig. S8a), reflecting their electron-like nature. Momentum-dependent dispersions of the TDSSs are shown in Fig. S8b. The Dirac point of TDSSs is clearly identified at the $E_B$ around 260 meV (Figs. S8a and S8b). In addition, photon energy-dependent measurement was also performed on the K-dosed GdSn₂ surface. The ARPES spectrum along the Γ - A direction reveals a non-dispersive feature at the $E_B$ of the Dirac point, exhibiting the intrinsic two-dimensional nature the TDSSs. Remarkably, the lower branch of the TDSSs merges into the bulk bands [see Fig. 4D(iii) in the main text, and the red box selected region in Fig. S8d], and only the upper branch crosses the $E_F$, which reverses the chirality of the spin texture at the Fermi surfaces, as we demonstrate further in the section S8.

Fig. S9 | Momentum dependence and photon energy dependence of the doped TDSSs in GdV₆Sn₆. a Constant energy contours at binding energies. b Momentum-dependent dispersions of the TDSSs. The corresponding momentum paths are indicated by the red lines in (a). c ARPES spectral intensity as a
function of photon energy along the $\Gamma - A$ direction. The momentum direction of the spectrum is marked by the yellow arrow in Fig. S6(c). d ARPES spectra taken along the $\Gamma - K - M$ direction, measured with 86 eV $\hat{C}$ polarization. All data were collected on the K-dosed GdSn$_2$ layer, at 20 K.
S10. The tunability of the VHS on the kagome termination

In situ alkali metal deposition has provided an effective way to tune surface doping in quantum materials like cuprates [38], iridates [39], iron-based superconductors [40], and topology materials [12]. To tune the novel electronic states in GdV₆Sn₆, we employ surface potassium deposition technique to elevate carrier density. As shown in Fig. 4 in the main text, we successfully realize manipulation of the TDSSs and VHSs on the Gd termination.

Figure S10 presents doping dependence of the electronic structure recorded on the kagome termination. Upon surface doping, the hole-like bulk band is doped from slightly above the Fermi level ($E_F$) to below $E_F$ [see the red dashed curve in Figs. S10a(i) and b(i)]. Meanwhile, the bottom of the electron-like VHS1 band drops [highlighted by the red dashed curve in Figs. 10a(ii) and b(ii)], which is further accentuated by the EDCs taken on the VHS1 band (Fig. S10c). The downshifts of the bulk band collected on the kagome termination (Fig. S10) are in line with those of the Gd termination (Figs. 4E, 4F and 4H in the main text), consistently demonstrating the tunability of the VHS in GdV₆Sn₆.

**Fig. S10** | Manipulation of the VHS states on the kagome-termination via *in-situ* potassium deposition.
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**a** ARPES spectra taken along the $\overline{\Gamma}$ - $\overline{K}$ direction, on the pristine surface (V-termination), measured with 76 eV LH (i) and linear horizontal (LV) (ii) polarizations. Red dashed curve in (i) highlights the bulk band BS1. The arrow in (ii) indicates the VHS1 band. **b** Same as (a), but measured on the K dosed surface. **c** Doping evolution of the EDC extracted at the $\overline{M}$ point, measured with LV polarization, as indicated by the red line in [a(ii)].
S11. Electronic structure of GdV₆Sn₆ in ferromagnetism

Figure S11 shows the electronic structure in the ferromagnetic state. The magnetic moment of Gd atom is about 7 μB. As the states near the Fermi level is dominated by V d orbitals, the magnetic splitting originating from the coupling between V kagome and Gd layers is relatively small but visible.

Fig. S11|DFT calculated electronic structure of GdV₆Sn₆ in ferromagnetism.
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