Doping evolution of superconductivity, charge order and band topology in hole-doped topological kagome superconductors $\text{Cs}(\text{V}_{1-x}\text{Ti}_x)\text{Sb}_5$ 

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The newly discovered Kagome superconductors $A\text{V}_3\text{Sb}_5$ ($A=\text{K}, \text{Rb}, \text{Cs}$) exhibit superconductivity, charge order, and band topology simultaneously. To explore the intricate interplay between the superconducting and charge orders, we investigate the doping evolution of superconductivity, charge-density-wave (CDW) order and band topology in doped topological kagome superconductors $\text{Cs}(\text{V}_{1-x}\text{Ti}_x)\text{Sb}_5$ where the Ti-dopant introduces hole-like charge carriers. Despite the absence of the CDW phase transition in doped compounds even for the lowest doping level of $x = 0.047$, the superconductivity survives in all doped samples with enhanced critical temperatures. The high-resolution angle-resolved photoemission spectroscopy (ARPES) measurements reveal that the Ti-dopant in the kagome plane lowers the chemical potential, pushing the van Hove singularity (VHS) at $M$ point above the Fermi level. First-principle simulations corroborate the doping evolution of the band structure observed in ARPES, and affirm that the CDW instability does not occur once the VHS is pushed above the Fermi level, explaining the absence of the CDW ordering in our doped samples $\text{Cs}(\text{V}_{1-x}\text{Ti}_x)\text{Sb}_5$. Our results demonstrate a competition between the CDW and superconducting orders in the kagome-metal superconductor $\text{CsV}_3\text{Sb}_5$, although the superconductivity is likely inconsequential of the CDW order.

The transition-metal kagome compounds host novel correlated and topological electronic states and have been catching lots of attention in recent years [1–11]. Due to the geometrical frustration for electron hoppings, the simple kagomé lattice band structure possesses Dirac cones, flat band, and saddle point with van Hove singularities (VHS) in the electronic density of states (DOS), facilitating the emergence of various electronic orders [1–3, 12–14]. Among the kagome metal compounds, the charge-density-wave (CDW) charge order and superconductivity arise successively at around 78-103 K and 0.92-2.5 K, respectively, and intertwine with various electronic orders [1–3, 12–14]. The CDW order can be completely suppressed under low pressures with emerging the superconducting domes [35–37], implying a competition between the CDW order and the superconductivity [38]. The fragility of CDW against pressures suggests that the electronic structure determines its nature, however, no ARPES measurement of the band structure in response to external perturbations has been reported.

In this letter, we substitute vanadium with titanium in $\text{CsV}_3\text{Sb}_5$ to modulate the chemical potential and study the doping evolution of superconductivity, charge order and band topology in $\text{Cs}(\text{V}_{1-x}\text{Ti}_x)\text{Sb}_5$. We do not observe the CDW phase transition in the doped compounds even for the doping level of $x = 0.047$ in the transport and thermodynamic measurements. Meanwhile, the superconductivity remains with an enhanced temperature in all doped samples. From the ARPES measurements, we reveal that the overall pattern of band structures remains unchanged upon doping, but the Fermi level lowers down and hence the VHS1 at the $M$ point (Fig. 3) rises up in $\text{Cs}(\text{V}_{1-x}\text{Ti}_x)\text{Sb}_5$. Our first-principles simulations confirm the band evolution upon doping and affirm that the CDW instability in the parent compound [39] does not occur once VHS1 is pushed above the Fermi level, accounting for the absence of CDW in the doped samples. Except for the parent compound, the superconducting temperature $T_c$ decreases with increasing the doping levels, coinciding with the DOS of...
FIG. 1. Thermodynamic and transport properties of Cs(V_{1-x}Ti_x)_3Sb_5. (a) Temperature-dependent uniform magnetic susceptibilities $\chi = M/H$ with the c-axis directional field $H = 10^4$ Oe. (b) The zero-field-cool (ZFC) and field-cool (FC) magnetic susceptibility data under $H = 20$ Oe. (c) Temperature-dependent specific heat coefficient $C_p/T$ at zero field. The inset is the zoom-in low temperature data. (d) Temperature-dependent resistivity $\rho$ with respect to the 300 K result. (e) The zoom-in low temperature data of (d). (f) The doping dependent CDW and superconducting transition temperatures.

the Sb-derived electron pocket at $\Gamma$ point. The parent compound has a larger DOS of the Sb electron pocket, but a lower $T_c$ than the doped samples, indicating a competition between the CDW order and the superconductivity.

Cs(V_{1-x}Ti_x)_3Sb_5 single crystals with the titanium doping were synthesized by the self-flux method with the starting ratio Cs : V : Ti : Sb = 2 : 1 : x : 6 for $0 \leq x \leq 0.5$ [40]. The chemical composition of Cs(V_{1-x}Ti_x)_3Sb_5 was identified by Energy Dispersive X-Ray Spectroscopy (EDX, Phenom ProX). Fig. 1 summarizes the thermodynamic and transport properties of Cs(V_{1-x}Ti_x)_3Sb_5 with different doping levels $x$. Titanium sits on the left next to vanadium, and the substitution reduces the total number of charge carriers. The lowest doping concentration with $x = 0.047$ of our samples already completely suppresses the CDW transition. However, the superconducting state at low temperatures survives at all doping levels ($0.047 \leq x \leq 0.117$) with enhanced critical temperatures. In Fig. 1(a), we compare the temperature-dependent magnetic susceptibilities of the parent compound CsV_3Sb_5 and the Ti-doped ones Cs(V_{1-x}Ti_x)_3Sb_5. A sudden drop of the magnetization in CsV_3Sb_5 appears at 94 K, indicating the CDW transition [9]. For all Ti-doped samples, no CDW anomaly occurs, implying the absence of the CDW transition. We observe increasingly weakened magnetization signature at both high and low temperatures with the increasing doping levels $x$, suggesting a reduction of DOS of itinerant electrons or local correlations for vanadium 3d electrons upon doping. The complete suppression of CDW is also revealed in the heat capacity and the resistivity measurements in Figs. 1(c) and (d)-(e). All the samples of Cs(V_{1-x}Ti_x)_3Sb_5 display the superconducting transition at low temperatures in the magnetization (Fig. 1(b)), heat capacity (Fig. 1(c)) and resistivity (Fig. 1(d)-(e)) measurements. Fig. 1(f) is the doping-dependent transition temperatures of the CDW order and the superconductivity.

High-resolution ARPES measurements are performed to investigate the band structure evolution of the Cs(V_{1-x}Ti_x)_3Sb_5 with different doping level. As detailed photon energy dependent ARPES measurements have been performed [24], the value of $k_z$ can be determined precisely. Fig. 2(a) shows band structures measured at 17 K of different doping level with 31 eV photons, which correspond to the A-H-L plane ($k_z = \pi$). As we can see, an electron-like band around A, several Dirac cone-like dispersions along the A-H-L direction can be clearly resolved in this system. To focus on how these bands evolve with different doping, we extract the their band dispersions by fitting the momentum distribution curves (MDCs), marked by A-D in different colors, and then draw them in Fig. 2(b) separately. The overall band structures shift upwards with different doping level monotonously, in accordance with the fact that hole doping pushes the Fermi level ($E_F$) downwards. By substituting 11% percent of V atoms with Ti atoms, the band structure can be shifted up about 100 meV. Our results prove that the substitution of Ti is an efficient way to tune the Fermi level of CsV_3Sb_5 system.

As shown in Fig. 1, there is no CDW anomaly for all the
Ti-doped CsV₃Sb₅ compounds. Since the VHSs at the M point near the Fermi level play crucial roles in the CDW instability, it is of importance to figure out how the VHSs near the Fermi level evolve with different doping. We select three typical doping levels (x = 0, 0.047 and 0.11) and analyse their detailed band structures around the L point, as shown in Figs. 3(a-c). According to the previous ARPES study [24], the ARPES spectral intensity is dominated by contribution from only the top two layers, resulting in the lack of clear kₚ dispersion along M-L and the missing information of the VHSs around the M point. To have a fully clear understanding on the evolutions of band structure and VHSs upon doping, first-principles simulations within density-functional theory (DFT) are performed here [40]. As revealed by ARPES measurements in Fig. 2, except for the overall upward shift, all the features of band structure remains almost unchanged under doping, meaning that the Ti-dopant doesn’t break any crystal symmetry of CsV₃Sb₅. Therefore, we employ a virtual crystal approximation in the simulations of Ti-substituted V in CsV₃Sb₅, which is qualitatively consistent with the results calculated using the supercell of Cs(V₁₋ₓTiₓ)₃Sb₅ [40].

Fig. 3(a) demonstrates that the DFT calculated band structure can well fit the ARPES measurements of CsV₃Sb₅. As shown in Fig. 3(d), upon doping of Ti, the overall band structures are shifted upwards, consistent with the hole-doped effect revealed by ARPES. Now we focus on the evolutions of VHSs. Around the Fermi level, there are two VHSs (VHS1 and VHS2) at both the M and L points, as marked by red arrows in Fig. 3(d). It is noted that the VHS1 has a large kₚ dispersion along M-L, and it crosses the Fermi level from the M (below E_F) to L (above E_F) points, as shown in Fig. 3(e). While the VHS2 has almost no kₚ dispersion along M-L. In addition, the VHS1 has a higher density of states (DOS) near E_F, which accounts for the CDW instability as will be demonstrated below.

Around the L point, the VHS1 cannot be measured by ARPES due to E(VHS1) > E_F, but the VHS2 is observed under all our doping levels, as shown in Figs. 3(a-c). We extract the band dispersions around VHS2 [red circles in Figs. 3(a-c)] and fit them with red dashed lines. Thus the exact positions of VHS2 can be obtained quantitatively. In Fig. 3(f), we summarize the calculated and experimental energy shifts of these two VHSs under different doping levels where both the energies of VHS1 at M and L are plotted here. In general, all the energies of VHSs are shifted upward with respect to E_F as increasing doping level. The calculated energy evolution of VHS2 [green circles in Fig. 3(f)] is in good agreement with the experimental results [blue stars in Fig. 3(f)]. Focusing on the VHS1, its kₚ dispersion along M-L allows the VHS1 to be located at any energy position between the energies of the M and L points, which has been plotted by the cyan region in Fig. 3(f). In other words, the VHS1 can be in principle located at any position of the cyan region except for some energy gaps caused by the coupling between VHS1 and other bands. It is noted that the CDW instability is mainly induced by the VHS at E_F. Therefore, that the Fermi energy is included in the range of VHS1 energy between the M and L points is a necessary condition for the formation of CDW. As shown in Fig. 3(f), for
FIG. 3. Doping evolution of VHSs at the M/L points in Cs(V$_{1-x}$Ti$_x$)$_3$Sb$_5$. (a)-(c) ARPES measurements along the H-L-H direction with selected doping levels. In (a) the DFT calculated band structure (green dashed lines) is also over-plotted. The DFT calculated projected bulk band structure along $\Gamma$-M-K-$\Gamma$ and A-L-H-A directions in (d) and along the M-L direction in (e). (f) The comparison between the calculated and experimental results on the energy shift of VHSs with doping. (g-h) The calculated phonon dispersion relations with doping level $x = 0$ (g) and $x = 0.07$ (h).

The very different doping behaviors in Cs(V$_{1-x}$Ti$_x$)$_3$Sb$_5$ suggest that the CDW and the superconducting orders in the parent compound have different underlying electronic origins. Our results affirm the VHS saddle point on the V bands leads to the CDW instability, consistent with the previous theory [39]. However, it is unlikely the reason for the superconductivity since $T_c$ survives even when the doping of Ti pushes VHS1 above the Fermi level, at odds with the previous speculation of the nature of pairing around the VHS [41]. Recent NMR and magnetic penetration depth measurements [42, 43] suggest a conventional $s$-wave superconducting paring in CsV$_3$Sb$_5$, which unlikely occurs on the V bands regarding the robustness of the superconductivity upon

FIG. 4. Doping evolution of the CDW order $T_{\text{cdw}}$, superconducting transition temperatures $T_c$ and effective mass $m^*$ of the Sb-derived electron pockets at $\Gamma$ of Cs(V$_{1-x}$Ti$_x$)$_3$Sb$_5$. 

the parent compound CsV$_3$Sb$_5$, the VHS1 at M (L) is below (above) $E_F$. With hole doping, the VHS1 at M shifts upward and is pushed above the Fermi level beyond 4.5% doping concentration ($x > 0.045$). As a consequence, we can divide the phase diagram by $x = 0.045$. When $x < 0.045$, the VHS1 can be located at the Fermi level, and the CDW order may be formed. When $x > 0.045$, all the energy of VHS1 is above the Fermi level, and the CDW order is hard to form. This result can well explain why no CDW anomaly occurs for all the Cs(V$_{1-x}$Ti$_x$)$_3$Sb$_5$ compounds with $x \geq 0.047$. As demonstrated by previous study [39], the CDW instability can be identified by the imaginary frequency of the phonon spectrum. Here, we compare the calculated phonon spectrum under different doping level with $x = 0$ and $x = 0.07$, as shown in Figs. 3(g,h). For the undoped CsV$_3$Sb$_5$, unstable modes with imaginary frequencies exist at M and L. For the 7% doping concentration, these unstable modes disappear, which implies the absence of the CDW order. Combining all the results above, it can be concluded that the hole doping pushes the VHS1 above the Fermi level, and then lead to the disappearance of the CDW instabilities. Our results also reveal that the VHS1 plays a dominant role in the formation of the CDW order.
doping in Cs(V_{1-x}Ti_x)Sb_5. To investigate the relationship between the superconductivity and the Sb electron pocket, we compare the effective mass \( m^* \) of the Sb electron pocket at the \( \Gamma \) point (C3 band in Fig. 2) with the superconducting transition temperatures \( T_c \). Due to the quasi-2D structure, \( m^* \) is proportional to DOS, and its doping evolution agrees well with \( T_c \) in \( Cs(V_{1-x}Ti_x)Sb_5 \) except the parent compound, as shown in Fig. 4. The similar doping behaviors of \( m^* \) and \( T_c \) in the doped samples imply a BCS scenario, consistent with the \( s \)-wave paring. The parent compound has the largest effective mass \( m^* \), but with a suppressed \( T_c \), consistent with the competition relationship between the CDW order and the superconductivity [15, 16, 35–38].

In conclusion, the doping evolution of the band structures in \( Cs(V_{1-x}Ti_x)Sb_5 \) manifests that the van Hove singularity is the origin of the CDW ordering, but not of the superconductivity which likely occurs on the Sb electron pocket. Although they have different origins, the charge order and the superconductivity displays a competition relation, accounting for the intertwined orders in the parent compound CsV_2Sb_5.

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