Large kagome family candidates with topological superconductivity and charge density waves

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A group of newly discovered non-magnetic metal kagome structures AV3Sb5 (A = K, Rb, Cs) have aroused widespread interest in experiment and theory due to their unusual charge density wave (CDW) and intertwined superconductivity. However, they all possess weak electron-phonon coupling (EPC) and low superconducting transition temperature \( T_c \). Here, we performed high-throughput first-principles calculations on novel kagome candidates with AV3Sb5 prototype structure, and proposed 24 dynamically novel stable kagome metals. The calculation based on Bardeen-Cooper-Schrieffer theory shows that most of these metals are superconductors with much stronger EPC than the reported AV3Sb5 materials, and their \( T_c \) are between 0.3 and 5.0K. Additionally, several compounds, such as KZr3Pb5 with the highest \( T_c \), are identified as \( \mathbb{Z}_2 \) topological metals with clear Dirac cone topological surface states near Fermi level. And NaZr2As5 is shown to have possible CDW phases. Our results provide rich platforms for exploring various new physics with kagome structure, in which the coexistence of superconductivity and nontrivial topological nature provides promising insights for the discovery of topological superconductors.

Introduction.—In 2019, a new class of nonmagnetic metals AV3Sb5 (A = K, Rb, Cs) with perfect vanadium kagome net were synthesized [1]. Since then, surprises have emerged in the study of these structures. The electronic structures of AV3Sb5 show Dirac nodal lines, nontrivial \( \mathbb{Z}_2 \) topological band indices and clear topological surface states near Fermi level, indicating that their normal states are \( \mathbb{Z}_2 \) topological metals [1–4]. The superconducting transition temperatures \( T_c \) of KV3Sb5, RbV3Sb5 and CsV3Sb5 are 0.93 [2], 0.92 [5] and 2.5K [3](2.3K [6, 7]), respectively, and the temperatures \( T^* \) corresponding charge density wave (CDW) transition are 78 [8], 102 [9] and 94K [10], respectively. The exotic CDW states of non electron-phonon coupling (EPC) mechanism [9] display many unconventional characteristics. The CDWs in these three compounds exhibit chiral anisotropy [11–13] and reduce the density of electronic states near the Fermi level [11, 14–16]. Various evidences, including giant anomalous Hall response [17, 18], CDW chirality adjustable by magnetic field [11–13], edge supercurrent [19] and spontaneous internal magnetic field [20], indicate that the charge order may break the time-reversal symmetry, which has also been verified theoretically [11, 21, 22]. Moreover, CsV3Sb5 samples have been found to own roton pair density wave [7], which is similar to the one in unconventional high-\( T_c \) cuprate superconductors [23]. The coexistence of \( V \)- and U-shaped superconducting gaps [24] and the anisotropic superconducting properties in CsV3Sb5 [25] imply the possible multi-band superconducting pairing. Intertwinned superconductivity with CDW shows many new features. For instance, \( T_c \) exhibits an unconventional double dome behavior, and \( T^* \) decreases rapidly with increasing pressure [10, 26, 27]. The second dome of \( T_c \) and the disappearance of \( T^* \) occur at the same pressure. The superconducting properties and charge order of AV3Sb5 can also be tuned by magnetic impurity [24], strain [28] and thickness [29–31], which dramatically enriches the phase diagram. The experimental and theoretical studies on AV3Sb5 show a complementary and rapid trend. However, to further explore the exotic properties of AV3Sb5, more candidate compounds based on the AV3Sb5 prototype structure are urgently needed.

In this paper, we first apply the high-throughput first-principles calculations to 800 new kagome structures based on AV3Sb5 prototype, and find 24 dynamically stable metal compounds. Then, we carefully study their superconducting and topological properties. The results show that 14 novel compounds are superconductors with the \( T_c \) between 0.3 and 5.0K. Moreover, several structures, including KZr3Pb5 with the highest \( T_c \), have strong \( \mathbb{Z}_2 \) indices with abundant nontrivial topological surface states near Fermi surface, suggesting that they are \( \mathbb{Z}_2 \) topological metals. The coexistence of superconductivity and nontrivial band topology opens a door for the discovery of topological superconductivity based on the kagome net. Additionally, we also find two possible CDW phases in NaZr2As5, which exhibit soft modes in phonon spectrum, and may provide useful information for further understanding the CDW phases in AV3Sb5.

Crystal structure of AV3Sb5.—The AV3Sb5 crystallize in a layered structure with the space group of P6/mmm (No.191) as shown in FIG. 1. The perfect V-kagome net mixed with the Sb-triangular net of is located in the middle layer, which is sandwiched by two additional honeycomb layers of Sb atoms. The upper and lower triangular layers of alkali metal A atoms have a large bond distance relative to the middle V-Sb layer and are loosely bonded.
TABLE I. Electronic density of states at Fermi energy $N(E_F)$ (eV$^{-1}$f.u.$^{-1}$), logarithmic average frequency $\omega_{ag}$ (K), EPC $\lambda(\omega = \infty)$ and $T_c$ of 14 stable compounds.

| Compound        | $N(E_F)$ | $\omega_{ag}$ (K) | $\lambda$ | $T_c$ (K) |
|-----------------|----------|--------------------|-----------|-----------|
| KNb$_3$Sn$_5$   | 4.22     | 149.1              | 0.52      | 2.102     |
| CsRu$_3$Ge$_5$  | 3.19     | 170.8              | 0.36      | 0.353     |
| RbTi$_3$Bi$_5$  | 5.96     | 149.8              | 0.41      | 0.719     |
| CsTi$_3$Bi$_5$  | 5.96     | 163.4              | 0.35      | 0.316     |
| KT$_3$Pb$_5$    | 7.29     | 157.9              | 0.51      | 2.039     |
| RbTi$_3$Pb$_5$  | 7.50     | 156.5              | 0.50      | 1.857     |
| KT$_3$Sn$_5$    | 6.57     | 180.3              | 0.42      | 0.974     |
| RbTi$_3$Sn$_5$  | 6.50     | 182.5              | 0.42      | 0.961     |
| CsTi$_3$Sn$_5$  | 6.87     | 174.6              | 0.45      | 1.375     |
| CsZ$_3$As$_5$   | 7.30     | 125.8              | 0.56      | 2.289     |
| KZ$_3$Pb$_5$    | 6.47     | 94.1               | 0.91      | 5.027     |
| RbZ$_3$Pb$_5$   | 6.56     | 111.9              | 0.72      | 4.154     |
| CsZ$_3$Pb$_5$   | 6.53     | 119.2              | 0.58      | 2.438     |
| CsZ$_3$Te$_5$   | 2.88     | 123.5              | 0.48      | 1.266     |

TABLE II. Electronic density of states at Fermi energy $N(E_F)$ (eV$^{-1}$f.u.$^{-1}$), experimental superconducting temperature $T_{c exp}$ and $T_c$ of pristine phase of AZr$_3$Pb$_5$.

| Compound        | $N(E_F)$ | $\lambda$ | $T_c$ (K) | $T_{c exp}$ (K) |
|-----------------|----------|-----------|-----------|-----------------|
| KV$_3$Sb$_5$    | 2.9      | 0.38      | 0.22      | 0.93            |
| RB$_3$Sb$_5$    | 2.33     | 0.32      | 0.05      | 0.92            |
| CS$_3$Sb$_5$    | 1.30     | 0.25      | 0.0008    | 2.5 (2.3)       |
| KZ$_3$Pb$_5$    | 6.47     | 0.91      | 5.027     | -               |
| RB$_3$Pb$_5$    | 6.56     | 0.72      | 4.154     | -               |
| CS$_3$Pb$_5$    | 6.53     | 0.58      | 2.438     | -               |

We choose AZr$_3$Pb$_5$ group with relative higher $T_c$ for further discussions. The phonon spectra, phonon density of states (PhDOS), $\alpha^2F(\omega)$ and $\lambda(\omega)$ of this group are plotted in FIG. 2. We can see that the phonon spectra of

FIG. 1. The crystal structure of AV$_3$Sb$_5$. 22 new stable AB$_3$C$_5$ members with the same crystal structure as AV$_3$Sb$_5$ are also indicated.

Searching new structures.—The high-throughput first-principles calculations are used to search for kagome topological superconductor candidates (as indicated in FIG. S1 in Supplementary Material (SM)). Based on the prototype structure of AV$_3$Sb$_5$, 800 new compounds are constructed by replacing A with alkali metal elements Li, Na, K, Rb and Cs, replacing V with all transition metal elements (Ge, As, Se, Sn, Sb, Te, Pb, and Bi). These new compounds will be abbreviated as AB$_3$C$_5$ below. For all these new compounds, we first carry out fully geometric relaxation and replacing Sb with its neighboring elements (Ge, As, Se, Sn, Sb, Te, Pb, and Bi). These new compounds, we first carry out fully geometric relaxation and then using the phonon spectra to determine the dynamic stability of these compounds to further demonstrate their stability, as listed in Table S1. Electronic structure calculations show that they are all metals similar to AV$_3$Sb$_5$. Furthermore, 22 members maintain the same crystal structure as AV$_3$Sb$_5$ after structural optimization (as listed in FIG. 1), except for CsRu$_3$Ge$_5$ and RbCr$_3$Te$_5$, whose triangles in the kagome nets are twisted, which changes their space group to P6$_3$cm.
three compounds are very similar. Careful comparison of their phonon spectra shows that the faint phonon softening at L point gradually becomes obvious from K to Rb to Cs. It can be seen from PhDOS that the contributions of Pb and Zr atoms to PhDOS are mainly distributed in the relatively low and high frequency regions with much prominent peaks, respectively, while the PhDOS of alkali metal atoms distributed in the medium frequency region are very small. The relatively low frequency (<3THz) phonons corresponding to the vibration modes of Pb account for more than half of the total EPC. The $T_c$ of the three compounds decreases with the gradual increase of the atomic number of alkali metals as shown in Table I. The gradual decrease of $T_c$ from K to Rb to Cs is due to the negligible contribution of alkali metal elements to the EPC, and the increase of atomic radius from K to Cs, resulting in the gradual increase of the lattice parameters, is equivalent to applying a negative pressure (tensile strain) to the lattice, which significantly reduce the parameters related to the lattice and weakens the EPC.

**Electronic band structure and topological property.**— We plot the electronic energy bands and density of states (DOS) with spin-orbit coupling (SOC) for KZr$_3$Pb$_5$ in FIG. 3(a). The electronic band structures of RbZr$_3$Pb$_5$ and CsZr$_3$Pb$_5$ are also given in FIGs. S21-22. The 3D Fermi surface (FS) of KZr$_3$Pb$_5$ and its 2D slice at $k_z=0$ and $\pi$ planes are drawn in FIGs. 3(b)-(d), which is obviously different from the FS of AV$_3$Sb$_5$ that exhibits strong 2D characteristics. Furthermore, we can see the obvious Fermi surface nesting with the nesting vector parallel to A-L and A-H in the $k_z=\pi$ slice. A Z$_2$=0 and (d) $\pi$ slices. Different colors of FS refers to different band indices consistent with (a), (e) Product of parity and Z$_2$ indices of bands near Fermi level. (f) The Brillouin zone with high symmetry paths indicated. Topological surface states along (g) Γ-M-Γ and (h) K-M-K paths on (001) plane for KZr$_3$Pb$_5$. 

FIG. 2. The phonon spectra, projected PhDOS, Eliashberg spectral function $\alpha^2 F(\omega)$, and cumulative frequency dependent EPC $\lambda(\omega)$ of (a) KZr$_3$Pb$_5$, (b) RbZr$_3$Pb$_5$, (c) CsZr$_3$Pb$_5$.

FIG. 3. (a) The electronic energy bands and density of states calculated with SOC for KZr$_3$Pb$_5$. (b) 3D FS of KZr$_3$Pb$_5$, and its 2D maps at (c) $k_z=0$ and (d) $\pi$ slices. Different colors of FS refers to different band indices consistent with (a). (e) Product of parity and Z$_2$ indices of bands near Fermi level. (f) The Brillouin zone with high symmetry paths indicated. Topological surface states along (g) Γ-M-Γ and (h) K-M-K paths on (001) plane for KZr$_3$Pb$_5$. 

FIG. 4. (a) The phonon spectrum in FIG. 4(d) shows an obvious
softening acoustic phonon modes at M and L points at
the boundaries of Brillouin zone, and the imaginary
frequency at L point is slightly larger than that at M point,
which is very similar to AV$_3$Sb$_5$ [15, 32]. The symmetry
analysis on AV$_3$Sb$_5$ indicates that the irreducible repre-
sentations of the imaginary mode at M and L points are
M$^\Gamma_7$ and L$^\Gamma_2$, respectively, which is also consistent with
the previous studies [15, 34]. However, similar analysis
shows that the irreducible representations of the imagi-
nary mode in NaZr$_3$As$_5$ are M$^\Gamma_5$ and L$^\Gamma_3$, which makes
NaZr$_3$As$_5$ exhibit completely different distortions from
AV$_3$Sb$_5$. In consideration of one L point, it gives the
phase as shown in FIG. 4(b). The soft mode at M point
makes corner-shared triangles in layers rotate around the
corner, while the soft mode at L point leads to the distor-
tion of adjacent layers with an additional $\pi$-shift. Clock-
wise and counterclockwise distortions will generate the
same structure. The combination of all three unequal L
points gives a similar phase as shown in FIG. 4(c), which
differs from FIG. 4(b) in that Zr atoms in the kagome
layers rotate around the center of triangles. These two
structures have Ibam (No.72) and P6/mcc (No.192) space
groups, respectively. Both of them reduce the rotation
symmetry of C$_5$ to C$_2$, but still retain the spatial inver-
sion symmetry.

Their phonon spectra in FIGs. 4(e) and (f) show dy-
namic stability with completely disappeared imaginary
frequency, so both of them are possible CDW phases of
NaZr$_3$As$_5$. We label them CDW I and CDW II, respec-
tively. Compared with the pristine phase, the displace-
ment values of Zr atoms in kagome layer in CDW I and
CDW II are 0.12 and 0.15Å, respectively. The total en-
ergy as a function of displacement of Zr atoms is shown
in FIGs. 4(h) and (i). The total energies of the two sta-
ble CDW phases are 11.3 and 25.1meV lower than that of
pristine structure, respectively.

Unfolded energy bands and DOS of CDW phases in
FIG. S30 show no clear changes compared with the pris-
tine phase except that some gaps are opened and the
saddle point at the L point moves closer to the Fermi
level. Saddle-point nesting in electronic structure is un-
likely the origin of the CDW order NaZr$_3$As$_5$. The real
CDW phase, its origin and the possible interplay between
charge order and superconductivity in NaZr$_3$As$_5$ deserve
future experimental exploration. Besides NaZr$_3$As$_5$, we
also plot those structures with obvious soft modes at high
symmetry paths that may have CDW phases in FIG. S31.

Discussion. —In addition to AZr$_3$Pb$_5$, the calculated
results of all other stable AB$_3$C$_5$ members are presented
in FIGs. S3-23. The new AV$_3$C$_5$ members are not only
structurally similar to AV$_3$Sb$_5$, but also inherit many at-
ttractive features, such as Van Hove singularities at high
symmetry points near the Fermi level, Dirac points at the
Fermi level, Dirac nodal lines, and strong 2D character-
istics of the phonon spectrum and FS, which are worthy
of further studies.

For all AB$_3$C$_5$ kagome families proposed in this paper
and the reported AV$_3$Sb$_5$, we hardly see some obvious
flat bands in band structures. To further interpret this
feature, we construct a tight-binding model in SM. By
tuning the hopping parameters, we find that with the in-
crease of the hopping parameters between B atoms in the
kagome lattice and C atoms, the flat band becomes more
dispersive as seen in FIG. S27. C atoms and kagome B
atoms are very close to each other, and the overlap of
their orbitals makes the interaction between them very
complex and destroys the destructive interference condi-
tion for the formation of a flat band in kagome lattice,
resulting in the disappearance of the flat band.

An important feature of those predicted structures be-
yond AV$_3$Sb$_5$ is their much stronger EPC strengths.
The calculated $T_c$ of KV$_3$Sb$_5$, RbV$_3$Sb$_5$, and CsV$_3$Sb$_5$ based
on BCS theory are 0.0008, 0.05 and 0.22K, respectively
[32], which are much lower than their experimental val-
ues (see Table II), because the CDW in AV$_3$Sb$_5$ reduces
the DOS near Fermi level and suppresses BCS supercon-
ductivity. This indicates there may be an unconventional
superconducting mechanism. This mechanism is also ex-
pected to appear in the materials listed in Table I. From
Table II, it can be observed that the calculated $T_c$ of
AZr$_3$Pb$_5$ are much higher than those of AV$_3$Sb$_5$, thereby
experimental $T_c$ of AZr$_3$Pb$_5$ may be higher.

The coexistence of superconductivity and topologi-
cal nontrivial surface states is essentially rare [35–38].
It is reported that the robust zero-bias conductance
peak in CsV$_3$Sb$_5$ exhibits similar characteristics to the

FIG. 4. Crystal structures of NaZr$_3$As$_5$ in the (a) 2 × 2 su-
percell of pristine phase, (b) CDW I phase, and (c) the CDW
II phase and their corresponding phonon spectra (d), (e) and
(f), respectively. (g) 3D FS of NaZr$_3$As$_5$. The comparision
of total energies $\Delta E$ for (b) pristine phase and CDW I, (i)
pristine phase and CDW II, where the distortion represents
the displacement of Zr atoms and $\Delta E$ stands for the relative
total energy with respect to the pristine phase per cell with
72 atoms. The Brillouin zone of (b) and high symmetry paths
of (e) are plotted in FIG. S30(d).
Bi$_2$Te$_3$/NbSe$_2$ heterostructures with Majorana bound state [6]. Our new compounds with both the superconducting ground state and the nontrivial topological surface states near the Fermi surface would provide a rich platform for exploring topological superconductivity and Majorana zero-energy modes.

Mature experimental methods like flux method have been used to synthesize high-quality and stable AV$_3$Sb$_5$ compounds, which is a prerequisite for the rapid development of experimental analysis. In the initial work of Brenden et al. for the AV$_3$Sb$_5$ family, they explore the combination of (K, Rb, Cs)(V, Nb, Ta)(Sb, Bi) under different synthetic conditions [1]. However, only KV$_3$Sb$_5$, RbV$_3$Sb$_5$, and CsV$_3$Sb$_5$ are crystallized. In this work, 800 AB$_3$C$_5$ members in the high-throughput screening process contain most of the combinations they explored. Our calculation results show that these compounds not synthesized in their experiment are dynamically unstable except KNb$_3$Sb$_5$. The agreement with the experimental results indicates that our present calculations are resolvable, and the stable structures presented here are very likely to be synthesized in future experiments. Very recently, a newly discovered family of kagome metals RV$_6$Sb$_6$ (R = Gd, Ho, Y) with two V-derived kagome layers in the primitive cell was also synthesized by flux method [39, 40]. Therefore, the versatile and matured flux method may be employed to synthesize the stable structures in Table S1.

Summary.—In conclusion, we calculate 800 new kagome candidates based on the prototype structure of AV$_3$Sb$_5$ using a high-throughput DFT screening process, and discover 24 dynamically novel stable metal compounds, including one ferromagnetic, one antiferromagnetic and 22 nonmagnetic structures. These compounds display many appealing properties similar to AV$_3$Sb$_5$. Furthermore, based on the McMillan-Allen Dynes approach, 14 compounds among them are predicted to be phonon-mediated BCS superconductors with $T_c$ between 0.3-5K. KZr$_3$Pb$_5$ with the highest $T_c$ exhibits strong $Z_2$ invariants of the energy bands and abundant nontrivial topological surface states near the Fermi level, revealing that it is a Z$_2$ topological metal. In addition, we also find two possible CDW phases in NaZr$_3$As$_5$. This present work would give more insights on the exploration of possible topological superconductors.

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