

Tunable magnetism and electron correlation in titanium-based Kagome metals RETi_3Bi_4 (RE = Yb, Pr, and Nd) by rare-earth engineering

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Article

Keywords:

Posted Date: October 2nd, 2023

DOI: <https://doi.org/10.21203/rs.3.rs-3318771/v1>

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Additional Declarations: There is **NO** Competing Interest.

Version of Record: A version of this preprint was published at Communications Materials on May 10th, 2024. See the published version at <https://doi.org/10.1038/s43246-024-00513-4>.

Abstract

Rare-earth engineering is an effective way to introduce and tune the magnetism in topological Kagome magnets, which has been acting as a fertile platform to investigate the quantum interactions between geometry, topology, spin, and correlation. Here we report the structure and properties of three newly discovered titanium-based Kagome metals RETi_3Bi_4 (RE = Yb, Pr, and Nd) with various magnetic states. They all crystalize in the orthogonal space group $Fmmm$ (No. 69), where slightly distorted Ti Kagome lattice, RE triangular lattice, Bi honeycomb and triangular lattices stack along the a axis. By changing the rare earth atoms on RE zag-zig chains, the magnetism can be tuned from nonmagnetic YbTi_3Bi_4 to short-range ordered PrTi_3Bi_4 ($T_{\text{anomaly}} \sim 8.2$ K), and finally to ferromagnetic NdTi_3Bi_4 ($T_{\text{c}} \sim 8.5$ K). The resistivity and specific heat capacity measurements demonstrate an evolution of electron correlation and density of states near the Fermi level with different rare earth atoms. *In-situ* resistance measurements of NdTi_3Bi_4 under high pressure further reveal a potential relationship between the electron correlation and ferromagnetic ordering temperature. These results highlight RETi_3Bi_4 as a new family of topological Kagome magnets to explore nontrivial band topology and exotic phases in Kagome materials.

1 Introduction

Topological Kagome magnets and superconductors have attracted tremendous attention in the past decades, which have been serving as a fundamental platform to investigate the quantum interactions between geometry, topology, spin, and correlation [1]. From Heisenberg's view [2], magnetic ions on the Kagome lattice with antiferromagnetic interaction would exhibit geometrical frustration [3], resulting in possible quantum-spin-liquid states [4], fractionalized excitations [5], and the absence of ordinary magnetic order [6, 7]. When focusing on the electronic structure of Kagome lattices with magnetic ions, the strong electron correlation originated from the flat band would stabilize a ferromagnetic (FM) ground state [8]. Besides, with Dirac fermions and van Hove singularities in its electronic structure, Kagome lattices would also show nontrivial topological properties [9, 10] and multiple long-range orders with instabilities of Fermi surface [11]. By introducing the magnetic degrees of freedom, correlated topological band structures can be realized in Kagome lattices with magnetic ions, known as topological Kagome magnets. Theoretically, the inclusion of spin-orbit coupling in the Kagome lattice would open gaps at the Dirac point and the touching point connecting flat band and quadratic band, making nonmagnetic Kagome lattice a quantum spin Hall insulator with nontrivial Z_2 topological invariants [12, 13]. Thus, Kagome lattice with magnetic ions showing out-of-plane FM ordering would be a Chern insulator with chiral edge states, which usually exhibits exotic quantum properties such as quantized anomalous Hall effect [14] and giant anomalous Hall conductance [15, 16]. Moreover, the large in-plane magnetization would close the gaps in topological Kagome magnets, acting as a tuning parameter for topological phases [1]. Considering the strong or weak interlayer interaction, Weyl point [17–19] or three-dimensional quantum anomalous Hall effect [20] is anticipated to show up in topological Kagome magnets. Due to the exotic quantum properties, topological Kagome magnets have great potential

applications for next-generation electronic technology, and it is essential to explore new topological Kagome magnets and their properties.

Basically, there are two strategies to realize topological Kagome magnets, putting magnetic ions on the Kagome lattice or intercalating magnetic ions between stacked Kagome lattice. Plenty of topological Kagome magnets have been reported with magnetic transition metal ions on the Kagome lattice, such as Mn_3X ($\text{X} = \text{Sn}, \text{Ge}$) with Mn-Kagome lattice [21–23], $\text{Co}_3\text{Sn}_2\text{S}_2$ [24–26] and CoSn [27, 28] with Co-Kagome lattice, Fe_3Sn_2 [16, 29, 30] and FeSn [31–33] with Fe-Kagome lattice, most of which have strong in-plane magnetization. In REMn_6Sn_6 ($\text{RE} = \text{Gd-Tm}, \text{Lu}$) with Mn-Kagome lattice, various magnetic states can be realized by substitution of rare-earth atoms, named rare-earth engineering, which results in quite different quantum transport behaviors [34, 35]. In particular, TbMn_6Sn_6 shows an out-of-plane magnetization and stands out as a Chern insulator with zero-field anomalous Hall, anomalous Nernst, and anomalous thermal Hall effects [10, 35–37]. For REV_6Sn_6 with similar crystal structure whereas having nonmagnetic V-Kagome lattice, weak magnetic couplings among the local $4f$ moments of various rare-earth atoms in REV_6Sn_6 result in not only various magnetic orderings [38–40], but also topologically nontrivial Dirac surface states (GdV_6Sn_6 and HoV_6Sn_6 [41–43]), quantum oscillation (YV_6Sn_6 [42]), quantum critical behavior (YbV_6Sn_6 [44]), and charge density waves (ScV_6Sn_6 [45–47]). The close relationship between rare-earth magnetism and topological electronic structure highlights that rare-earth engineering is an effective way to tune the exotic topological phases [35, 48–50]. Recently, rare-earth engineering has been successfully applied in the AM_3X_5 ($\text{A} = \text{K}, \text{Rb}, \text{Cs}$; $\text{M} = \text{Ti}, \text{V}$; $\text{X} = \text{Sb}, \text{Bi}$) family with V/Ti-Kagome lattice [51, 52] having combined charge density wave states, superconductivity, orbital-selective nematic order or electronic nematicity, and nontrivial band topology [53–60]. By introducing RE atoms in VSb or TiBi layers, vanadium-based Kagome magnets emerge [61], showing a FM-like ground state in EuV_3Sb_4 . However, the titanium-based Kagome magnets are still rare, i.e. only LaTi_3Bi_4 , CeTi_3Bi_4 , and EuTi_3Bi_4 [62] are reported but without systematic investigation.

By using rare-earth engineering, we report the synthesis and physical properties of three newly discovered titanium-based Kagome metals RETi_3Bi_4 ($\text{RE} = \text{Yb}, \text{Pr}, \text{and Nd}$). RETi_3Bi_4 ($\text{RE} = \text{Yb}, \text{Pr}, \text{and Nd}$) all crystalize in the orthogonal space group $Fmmm$ with stacking layers composed of Ti-Kagome lattice, RE triangular lattice, Bi honeycomb and triangular lattices. Compared with AM_3X_5 , the Ti-Kagome lattice in RETi_3Bi_4 is slightly distorted and zig-zag RE chains are formed in REBi bilayers, making it possible to have various magnetic states with different RE atoms. The combined measurements of magnetism, resistivity, and specific heat capacity show an evolution from the nonmagnetic YbTi_3Bi_4 to short-range ordered PrTi_3Bi_4 with an anomaly around 8.2 K, and finally to FM NdTi_3Bi_4 with $T_c = 8.5$ K. The power-law fitting of resistivity at low temperature suggests an enhancement of electron correlation by changing RE atoms from Yb to Pr and Nd, which is consistent with the increasing density of states near the Fermi level according to the Debye model fitting of specific heat capacity. By applying pressure, the FM order is slightly suppressed with an increasing value of power in NdTi_3Bi_4 , showing potential relationship between the magnetism and electron correlation. Therefore, RETi_3Bi_4 ($\text{RE} = \text{Yb}, \text{Pr}, \text{and Nd}$) should be a

new family of topological Kagome magnets to investigate the interplay between topologically nontrivial feature, magnetism, and electron correlation.

2 Experimental

Single Crystal Growth. RETi_3Bi_4 (RE = Yb, Pr and Nd) single crystals were grown by a high-temperature solution method using Bi as flux. The as-received Yb/Pr/Nd ingot (Alfa, 99.9%) was cut into small pieces, then mixed with Ti powder (99.95%, Alfa Aesar) and Bi granules (99.999%, Sinopharm) with a molar ratio of Yb/Pr/Nd : Ti : Bi = 2 : 4 : 12 in a fritted alumina crucible set (Canfield crucible set) [63] and sealed in a fused-silica ampoule at vacuum. The ampoule was heated to 1073 K over 15 h, held at the temperature for 24 h, and then slowly cooled down to 873 K at a rate of 2 K/h. At 873 K, hexagonal-shaped, shiny-silver single crystals with size up to 5 mm $\times$ 5 mm $\times$ 0.5 mm were separated from the remaining liquid by centrifuging the ampoule. Considering the possible air sensitivity of the surface, all manipulations and specimen preparation for structure characterization and property measurements were handled in an argon-filled glove box.

Structure Characterization and Composition Analysis. X-ray diffraction data were obtained using a PANalytical X'Pert PRO diffractometer (Cu K_α radiation, $\lambda = 1.54178 \text{ \AA}$) operated at 40 kV voltage and 40 mA current with a graphite monochromator in a reflection mode ($2\theta = 5^\circ - 100^\circ$, step size = 0.017°). Indexing and Rietveld refinement were performed using the DICVOL91 and FULLPROF programs [64]. Single crystal X-ray diffraction (SCXRD) data were collected using a Bruker D8 VENTURE with Mo K_α radiation ($\lambda = 0.71073 \text{ \AA}$) at 280 K for PrTi_3Bi_4 and NdTi_3Bi_4 . The structure was solved using a direct method and refined with the Olex2 [65] and Jana2020 [66] package. Due to the air sensitivity and two-dimensional feature of YbTi_3Bi_4 , its crystal structure cannot be determined only by SCXRD. The focused ion beam (FIB) method was used to prepare thin specimens of YbTi_3Bi_4 with a thickness of $\sim 50 \text{ nm}$ for scanning transmission electron microscopy analysis. The crystal structure of YbTi_3Bi_4 was characterized by high-angle annular dark-field (HAADF) images obtained using a JEOL ARM-200F transmission electron microscope with double Cs correctors for the condenser lens and objective lens. The available spatial resolution is better than 78 picometers at 200 kV. The morphology and analyses of elements were characterized using a scanning electron microscope (SEM-4800, Hitachi) equipped with an electron microprobe analyzer for semiquantitative elemental analysis in energy-dispersive spectroscopy (EDS) mode. Three spots in three different locations were measured on each crystal using EDS.

Physical Property Measurements. Temperature-dependent magnetic susceptibility was measured using a vibrating sample magnetometer system (Quantum Design) under a magnetic field (0.5 T) parallel ($H // bc$) and normal ($H \perp bc$) to the bc plane using both the zero-field-cooling (ZFC) and field-cooling (FC) protocols. Only magnetic susceptibility in FC protocol was measured under a larger field (1 T), and the field-dependent magnetization curves were measured under the magnetic field up to 7 T parallel and perpendicular to the bc plane. The resistivity and heat capacity measurements were carried out using a physical property measurement system (Quantum Design, 9 T). The resistivity was measured using the

standard four-probe configuration with the applied current (about 2 mA) parallel to the bc plane. Heat capacity measurement was carried out at temperature ranging from 2.2 K to 200 K at high vacuum (0.01 μ bar). To protect the sample from air and moisture, thin film of N-type grease (~ 0.05 mg) was spread to cover the sample in an argon-filled glove box first and then the sample was mounted on the square plate of specialized heat capacity pucks in air.

In-situ high-pressure resistance measurement. The standard four-probe resistance measurement under high pressure for NdTi_3Bi_4 sample was carried out at high-pressure synergetic measurement station, synergetic extreme condition user facility. A Cu-Be alloy diamond-anvil cell with 300 μm diamond culet in diameter was used. For the sample chamber, a 100 μm diameter hole was drilled in a rhenium gasket with a c-BN insulating layer, which was then filled with KBr as the pressure transmitting medium. Long-stick shaped NdTi_3Bi_4 single crystal sample was loaded into the sample chamber in an argon-filled glove box. Four Pt electrodes were contacted to the sample in the four-probe configuration. The pressure of the sample chamber could be calibrated by ruby fluorescence at room temperature.

3 Results and Discussion

Unlike the hexagonal prototypes AM_3X_5 ($A = \text{K, Rb, Cs}$; $M = \text{Ti, V}$; $X = \text{Sb, Bi}$) [51, 52, 60], all RETi_3Bi_4 crystallize in the orthogonal space group $Fmmm$ (No.69) with $a = 24.9(4)$ $\text{\AA}$, $b = 10.3(4)$ $\text{\AA}$, $c = 5.9(2)$ $\text{\AA}$ for YbTi_3Bi_4 ; $a = 24.9668(114)$ $\text{\AA}$, $b = 10.3248(49)$ $\text{\AA}$, $c = 5.9125(26)$ $\text{\AA}$ for PrTi_3Bi_4 ; $a = 24.9523(87)$ $\text{\AA}$, $b = 10.3327(27)$ $\text{\AA}$, $c = 5.9009(18)$ $\text{\AA}$ for NdTi_3Bi_4 , and $\alpha = \beta = \gamma = 90^\circ$ (Table S1-S5). As shown in Fig. 1a, the crystal structure is composed of the alternating stacking of REBi, TiBi and Bi layers along the a axis. Viewed along the a axis, a Ti Kagome-like lattice, a RE triangular lattice, two Bi honeycomb-like lattices and a Bi triangular lattice can be further resolved (Fig. 1b). There are two distinct differences between the crystal structure of AM_3X_5 and RETi_3Bi_4 , i.e. (i) the increase of the a axis owing to the stacking of (ii) distorted TiBi layer with distorted Ti Kagome lattice. Compared with ATi_3Bi_5 , both the Ti Kagome and Bi triangular lattice of RETi_3Bi_4 show inequivalent bonds and are not strictly in the bc plane (Fig. 1c), resulting in both in-plane and out-of-plane distortion of TiBi layer. Similar to LnV_3Sb_4 ($\text{Ln} = \text{Yb, and Eu}$) [61], the bilayer of REBi plane forms a quasi-one-dimensional zig-zag chain of RE atoms (Fig. 1d), on which the magnetic states could be tuned by rare-earth engineering.

The crystal structure is further confirmed by the X-ray diffraction patterns, which show a strong preferential orientation of $(00l)$ ($l = \text{even integer}$) reflections (Fig. 2a). Based on the position ($\sim 14.3^\circ$) of (004) diffraction peaks, the distance between corresponding structural units along the a axis is determined to be about 25 $\text{\AA}$, close to the results of SCXRD. As shown in the insets of Fig. 2a, the as-grown single crystals of RETi_3Bi_4 are all plate-like flakes with shiny metal luster, indicating a clear quasi-two-dimensional feature. Figure 2b shows the enlarged (0010) diffraction peaks for RETi_3Bi_4 , where the peak position of PrTi_3Bi_4 shifts 0.08° to lower angle and NdTi_3Bi_4 shifts 0.06° to higher angle compared to that of YbTi_3Bi_4 , indicating the largest distance between the structural units for PrTi_3Bi_4 and the smallest one for NdTi_3Bi_4 . The different (0010) peak position corresponds well with the lattice parameter

derived from SCXRD, where $a(\text{NdTi}_3\text{Bi}_4) < a(\text{YbTi}_3\text{Bi}_4) < a(\text{PrTi}_3\text{Bi}_4)$. Considering the cationic radius $r(\text{Yb}^{3+}) = 0.0858 \text{ nm} < r(\text{Yb}^{2+}) = 0.093 \text{ nm} < r(\text{Nd}^{3+}) = 0.0995 \text{ nm} < r(\text{Pr}^{3+}) = 0.1013 \text{ nm}$, the abnormal enhancement of lattice parameter a in YbTi_3Bi_4 might be attributed to the weakened out-of-plane interaction between YbBi bilayer with Yb^{2+} ions rather than Yb^{3+} ions. As shown in the selected area electron diffraction (SAED) pattern and HAADF image of YbTi_3Bi_4 collected along the $[0-26]$ zone axis (Fig. 2c, d), the construction layers can be clearly resolved along the a axis. In particular, the Ti atoms and Bi atoms are not on the same straight line, further confirming the out-of-plane distortion of TiBi layers. The chemical composition is determined to be RE : Ti : Bi $\sim$ 1 : 3 : 4 with a homogenous distribution according to the result of EDS (Figure S1-S2 and Table S6-S8).

Figure 3a-c shows the magnetic susceptibility of RETi_3Bi_4 under field parallel ($H // bc$) and normal ($H \perp bc$) to the bc plane. The in-plane ($H // bc$) magnetic susceptibilities are larger than the out-of-plane ($H \perp bc$) magnetic susceptibilities, showing a quasi-two-dimensional feature of all RETi_3Bi_4 . For YbTi_3Bi_4 , the ZFC and FC curves under a smaller magnetic field (0.5 T) show no bifurcation above 2 K, hinting there is no magnetic transition. Though without bifurcation in the ZFC and FC curves under the smaller magnetic field (0.5 T) above 2 K, a saturation tendency appears at low temperature in the magnetic susceptibility of PrTi_3Bi_4 for both $H // bc$ and $H \perp bc$, possibly indicating a short-range ordering around $T_{\text{anomaly}} \sim 8.2 \text{ K}$ (Figure S3). By contrast, a clear bifurcation in the ZFC and FC curves can be resolved for NdTi_3Bi_4 , which corresponds to a possible FM ordering with transition temperature $T_c = 8.5 \text{ K}$.

All the magnetic susceptibility decreases quickly around 20 K and follows the Curie-Weiss law at higher temperature (black lines), $\chi = \chi_0 + C/(T - \theta)$, where χ_0 is the temperature-independent contribution including the diamagnetic contribution of the orbital magnetic moment, C the Curie constant, and θ the Curie temperature. The effective moment can be further calculated from the equation $\mu_{\text{eff}} = \sqrt{\frac{8C}{n}}$, where n is the number of magnetic atom. For YbTi_3Bi_4 , an almost zero Curie temperature ($\sim 0.5 \text{ K}$) and a small effective moment ($< 0.3 \mu_B/\text{Yb}$) were found for both $H // bc$ and $H \perp bc$, indicating a possible non-magnetic ground state with zero-spin ($0 \mu_B$) Yb^{2+} . For PrTi_3Bi_4 , a positive but close-to-zero Curie temperature ($\sim 2.5 \text{ K}$) was determined and the effective moment was calculated to be $3.67(3) \mu_B/\text{Pr}$ for $H // bc$, close to the theoretical moment of Pr^{3+} ($3.58 \mu_B$). The result may suggest weak magnetic exchange couplings between different Pr triangular bilayers along the a axis. Shifting the magnetic field configuration into $H \perp bc$, the large negative Curie temperature (-100 K) suggests strong antiferromagnetic interactions between moments in the bc plane, and the overestimated effective moment ($4.72 \mu_B/\text{Pr}$) should be attributed to the geometrical spin frustration of Pr triangular lattice. By substituting Pr with Nd, a small negative Curie temperature (-3.6 K) for $H // bc$ also suggests weak magnetic exchange couplings between different Nd triangular bilayers along the a axis. Whereas the reduced Curie temperature (-36.7 K) for $H \perp bc$ may correspond to a weakened antiferromagnetic interactions in the bc plane, which might result in the emergence of FM order in NdTi_3Bi_4 . The derived

effective moments ($\sim 3.6 \mu_B$) of NdTi_3Bi_4 are close to the theoretical value of Nd^{3+} ($3.62 \mu_B$). Details of the Curie-Weiss fittings can be seen in Table S9 and Figure S4.

The magnetization curves of YbTi_3Bi_4 and PrTi_3Bi_4 show positive slopes and approach saturation around 7 T without any hysteresis (Fig. 3d, e), suggesting no FM order or spin-glass state at least above 2 K. The saturation moment of YbTi_3Bi_4 is $0.016 \mu_B$ for $H // bc$ and $0.006 \mu_B$ for $H \perp bc$, further confirming the non-magnetic feature ($4f^{14}$, $\mu_{\text{sat}} = 0 \mu_B$). The saturation moment of PrTi_3Bi_4 is $1.620 \mu_B$ for $H // bc$ and $0.855 \mu_B$ for $H \perp bc$, of which the former one is close to the saturation moment of Pr^{3+} ($4f^2$, $\mu_{\text{sat}} = 2.0 \mu_B$). The large difference between the in-plane and out-of-plane saturation moment should be attributed to the large anisotropy originated from the quasi-two-dimensional structure. For NdTi_3Bi_4 , obvious hysteresis loops exist at 2 K and gradually vanish beyond T_c (Fig. 3f and Figure S5), further confirming the existence of FM order. The smaller in-plane coercive magnetic field ($H_{c//} \sim 500 \text{ Oe} \ll H_{c\perp} \sim 5000 \text{ Oe}$) and larger saturation moment ($\mu_{\text{sat-//}} \sim 1.426 \mu_B > \mu_{\text{sat-}\perp} \sim 0.587 \mu_B$) suggest that the easy plane is the bc plane. Thus, various magnetic ground states have been realized in titanium-based Kagome magnets RETi_3Bi_4 by rare earth engineering, which so far contains nonmagnetic (YbTi_3Bi_4), possibly short-range ordered (PrTi_3Bi_4), and FM (NdTi_3Bi_4) states.

Figure 4a shows the temperature-dependent in-plane resistivity of RETi_3Bi_4 , which monotonically decreases with decreasing temperature, showing a metallic-like behavior (See Figure S6 for the electronic structure of PrTi_3Bi_4 and NdTi_3Bi_4). The residual resistance ratios ($\text{RRR} = \rho_{300 \text{ K}} / \rho_{10 \text{ K}}$) are calculated to be 3.50 for YbTi_3Bi_4 , 2.15 for PrTi_3Bi_4 , and 3.70 for NdTi_3Bi_4 , hinting the good crystallinity of RETi_3Bi_4 single crystals. With current (2 mA) being applied in the bc plane, the resistivity of YbTi_3Bi_4 shows no obvious anomaly down to 2 K. Unlike YbTi_3Bi_4 , the in-plane resistivity of PrTi_3Bi_4 at low temperature shows an anomaly with a broaden peak (5.2 K $\sim$ 9.7 K) in its first derivative curve (Figure S7), which might be correlated with the possible short-range order. Due to the existence of FM order in NdTi_3Bi_4 , the resistivity shows a distinct drop around $T_c = 8.5 \text{ K}$ and then approaches a saturation at 2 K. Similar to AV_3Sb_5 or ATi_3Bi_5 , the resistivity of RETi_3Bi_4 below 50 K can be well fitted using Power law: $\rho = \rho_0 + AT^a$, where the value of power a is dependent on the dominant scattering mechanism (Table S10). Usually, a takes the value of 3/2 for diffusive electron motion caused by strong electron correlation [67], 2 for moderate electron-electron scattering [68, 69], known as the Fermi liquid behavior [70], 3 for dominant $s-d$ scattering or electron-magnon scattering [71–73], and 5 for electron-phonon coupling [74]. As shown in Fig. 4b, the value of power (a) is fitted to be 2.00(3) for YbTi_3Bi_4 , 1.64(3) for PrTi_3Bi_4 and 1.51(2) for NdTi_3Bi_4 , indicating a strengthening of electron correlation by changing rare earth atoms from Yb to Pr, and then to Nd. In particular, the resistivity of NdTi_3Bi_4 below T_c can also be fitted using Power law with $a = 5.35(6)$, showing the strong electron-phonon coupling below T_c . By applying magnetic field perpendicular to the bc plane, the resistivity of NdTi_3Bi_4 below T_c shows a slight enhancement up to 5 T with almost unchanged transition temperature (Fig. 4c), which is a typical characteristic of FM order.

Figure 4d shows the temperature-dependent specific heat capacity of RETi_3Bi_4 , where no anomaly in the entire measured temperature range, a broad anomaly around 8.2 K, and a sharp peak at $T_c = 8.5$ K are observed for YbTi_3Bi_4 , PrTi_3Bi_4 , and NdTi_3Bi_4 , respectively. All the specific heat capacity of RETi_3Bi_4 single crystals exceed the Dulong–Petit limit ($3NR \sim 200 \text{ J mol}^{-1} \text{ K}^{-1}$, black dashed line) at higher temperature, which is attributed to the more prominent phonon contribution at higher temperature of N-type grease used for protecting the samples [75–77]. For NdTi_3Bi_4 , the peak at 8.5 K is reversible by cooling ($T\downarrow$) and warming ($T\uparrow$), indicating no structural transition. The low-temperature (2 K – 5 K for YbTi_3Bi_4 , 12 K – 18 K for PrTi_3Bi_4 and NdTi_3Bi_4) specific heat capacity of RETi_3Bi_4 is fitted by the Debye model $C = \gamma T + \beta T^3$, where γT and βT^3 represent the contribution from the electron and lattice, respectively (Fig. 4e). The Sommerfeld coefficient γ proportional to the density of states (DOSs) around Fermi level (E_F) [78] ($\gamma \propto g(E_F)$) are determined to be 20.7(7) mJ K⁻² per formula for YbTi_3Bi_4 , 762.6(8.9) mJ K⁻² per formula for PrTi_3Bi_4 , and 809.6(7.5) mJ K⁻² per formula for NdTi_3Bi_4 . For the insulating character of N-type grease, γ correlated with the electron contribution will not be affected at low temperature. The large γ indicates large DOSs near E_F for both PrTi_3Bi_4 and NdTi_3Bi_4 , making it possible for them to show strong electron correlation, which is consistent with the results of resistivity. The λ -shaped peak of specific heat capacity for NdTi_3Bi_4 broadens and shifts toward higher temperature upon increasing the out-of-plane magnetic field (Fig. 4f), further confirming the FM order of NdTi_3Bi_4 . Taking the nonmagnetic YbTi_3Bi_4 as the reference of specific heat capacity contributed by electron and phonon, the contribution of magnon and magnetic entropy of PrTi_3Bi_4 and NdTi_3Bi_4 are calculated (Figure S8), suggesting possible Pr^{3+} and Nd^{3+} in PrTi_3Bi_4 and NdTi_3Bi_4 , respectively.

Upon applying hydrostatic pressure, multiple superconducting domes can be induced in AV_3Sb_5 , showing the competition between CDW and superconductivity [79–81]. To explore the possible superconductivity, *in situ* resistance measurements of NdTi_3Bi_4 in the pressure range of 1.5 GPa – 41.9 GPa were performed by using a diamond-anvil cell (Fig. 5a-b). Up to 10.2 GPa, a small drop related to FM order can be observed in resistance with a superconducting-like transition happened beyond 3.8 GPa (inset of Fig. 5a), where both the FM ordering temperature and superconducting transition temperature are suppressed as the pressure increases (Figure S9). By increasing the pressure beyond 10.2 GPa, the drop related to FM order cannot be resolved because the superconducting transition temperature suddenly steps higher (Fig. 5c). Until the pressure reaches 28.1 GPa, the drop related to FM order shows up again owing to the quicker suppression of superconducting transition. However, zero resistance is not observed in all measured curves, which may hint a non-intrinsic superconducting transition. By carefully evaluating the upper critical field (Figure S10), the superconducting signals should be attributed to three pressure-induced Bi phases (Bi-II, Bi-III, and Bi-V) [82], which might come from the remaining flux droplets or be induced by pressure in this Bi-rich compounds. Though having the superconducting side phase, the resistance of NdTi_3Bi_4 beyond FM ordering temperature (10 K – 45 K) still follows the Power law, $R = R_0 + AT^a$. Figure 5d and Table S11 show the pressure dependence of fitted parameters, where residual resistance (R_0) increases and correlation coefficient (A) decreases with the increasing pressure. In

particular, the value of power (α) increases from 1.51(2) to 1.82(3) when the pressure increases from 0 GPa to 32.4 GPa, showing a decreasing trend of electron correlation. Figure 5e-f summarizes the evolution of FM ordering temperature and Sommerfeld coefficient with the value of power, where the stronger electron correlation corresponds to a higher FM ordering temperature and higher DOSs near E_F . This result will direct us to find Kagome magnets with higher FM ordering temperature, hereby having stronger electron correlation and higher DOSs near E_F .

4 Conclusion

In summary, three newly discovered titanium-based Kagome metals RETi_3Bi_4 (RE = Yb, Pr, and Nd) are reported. Various magnetic states have been realized, including nonmagnetic YbTi_3Bi_4 , paramagnetic-like PrTi_3Bi_4 with an anomaly around $T_{\text{anomaly}} \sim 8.2$ K, and FM NdTi_3Bi_4 with $T_c = 8.5$ K. Both the electron correlation and DOSs around Fermi level are tuned by different rare earth atoms, indicating rare-earth engineering as an effective strategy to explore new materials and tune their exotic phases. This work extends the RETi_3Bi_4 family with diverse magnetic states a new platform to investigate the interplay between topologically nontrivial feature, magnetism, and electron correlation. Further calculations and experimental characterizations, like anisotropic magnetism or magnetoresistance, angle-resolved photoemission spectroscopy, and scanning tunneling microscopy or spectroscopy, are all desired to unravel the possible exotic topological phases of titanium-based Kagome metals RETi_3Bi_4 .

Declarations

Author information

Notes

The authors declare no competing financial interest.

Acknowledgements

L. Chen, Y. Zhou, H. Zhang, and X.C. Ji contributed equally to this work. L. Chen, Y. Zhou, and G. Wang would like to thank Prof. X. L. Chen, Prof. J. P. Hu, Prof. T. Qian at Institute of Physics, Chinese Academy of Sciences, and Prof. J. Ma at Shanghai Jiao Tong University for helpful discussions. This work was partially supported by the National Key Research and Development Program of China (Grant Nos. 2022YFA1403900 and 2018YFE0202600) and the National Natural Science Foundation of China (Grant No. 51832010 and 11888101).

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Figures

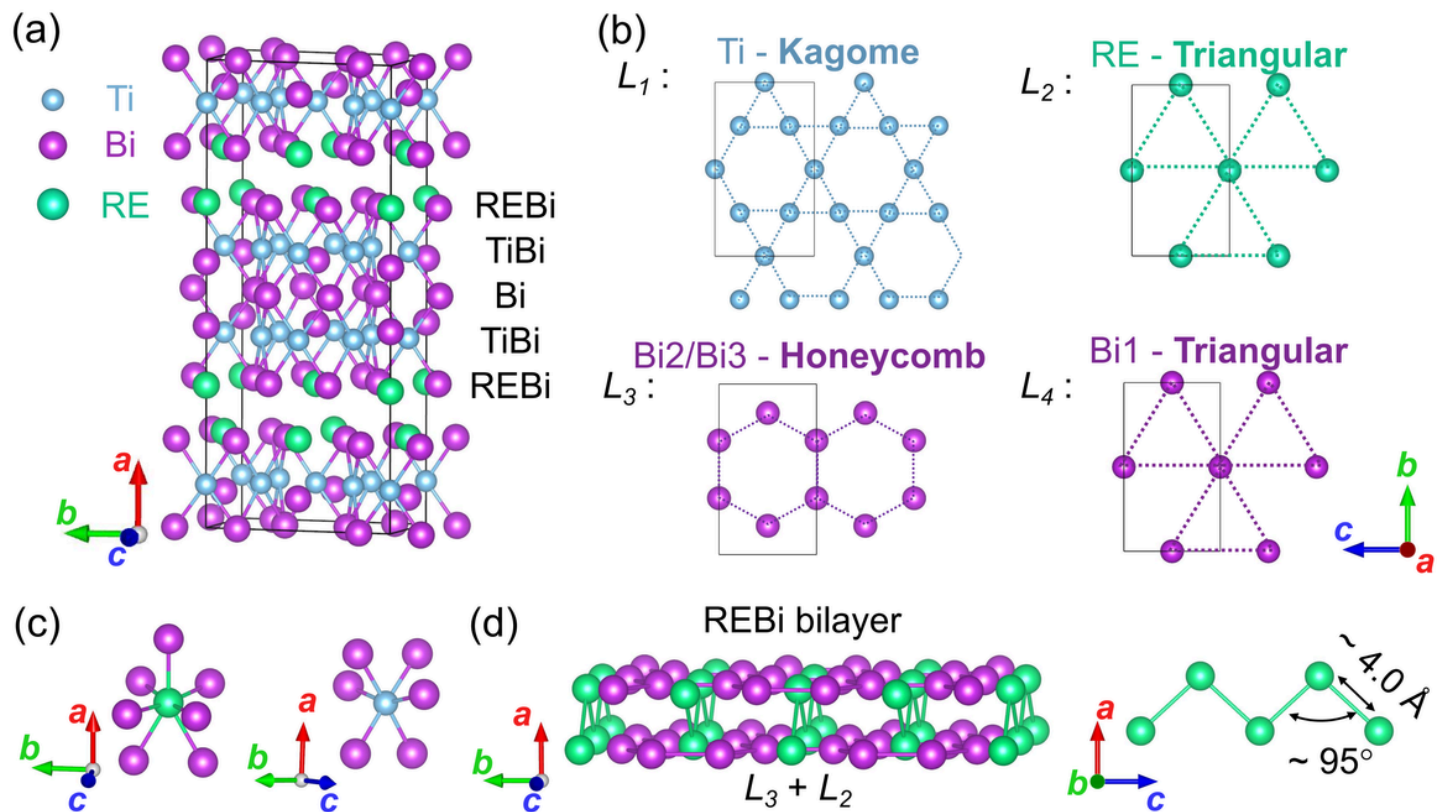


Figure 1

Crystal structure of RETi_3Bi_4 (RE = Yb, Pr and Nd). (a) Crystal structure of RETi_3Bi_4 (RE = Yb, Pr and Nd). (b) The sublattices viewed along the a axis, including Ti Kagome-like lattice (L_1), RE triangular lattice (L_2), Bi honeycomb-like lattices (L_3), and Bi triangular lattice (L_4). (c) The coordination environment of RE and Ti, respectively. (d) The REBi bilayer and zig-zag chain of RE atoms.

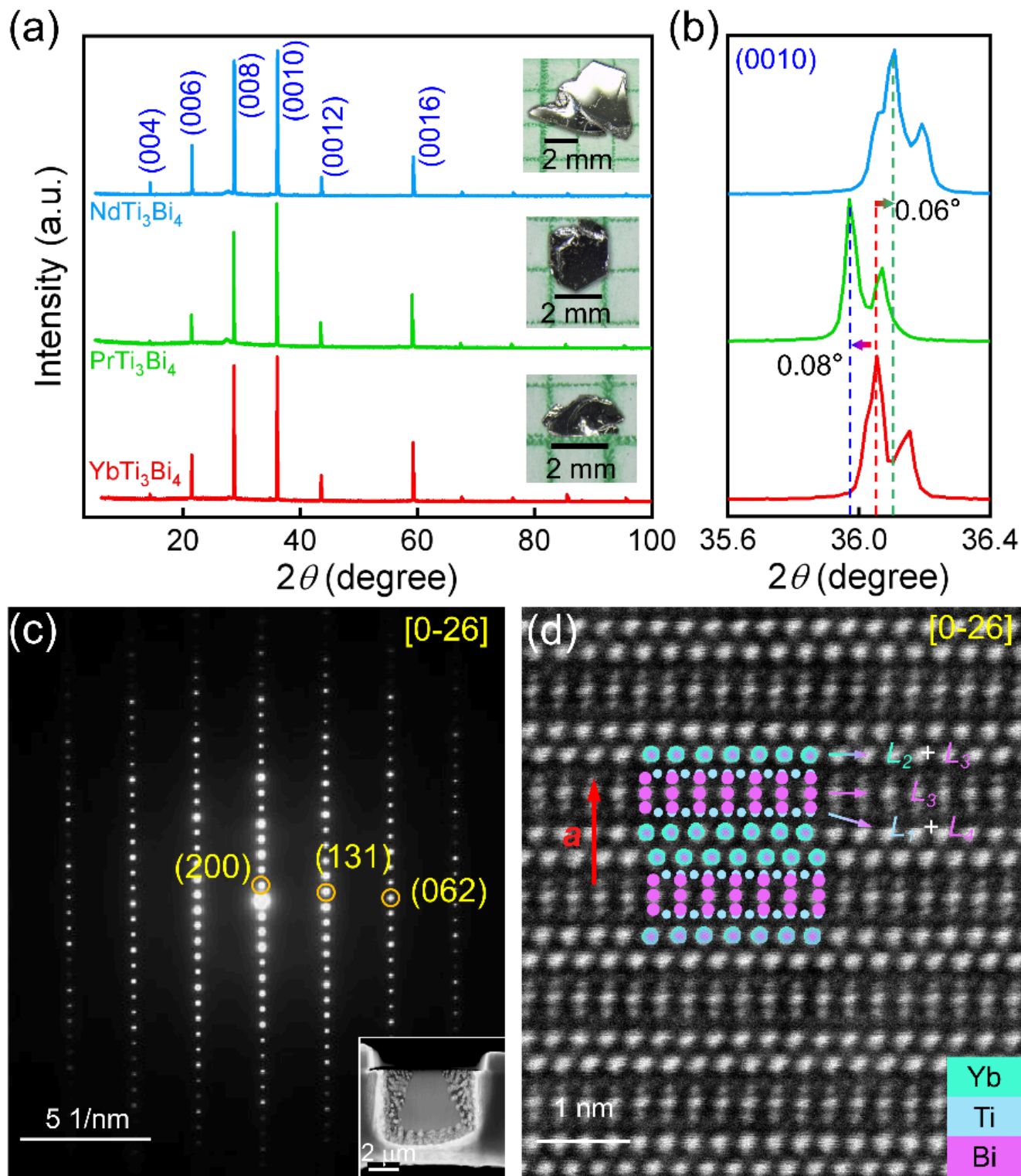


Figure 2

Single Crystals of RETi_3Bi_4 (RE = Yb, Pr and Nd). (a) X-ray diffraction patterns of as-grown RETi_3Bi_4 (RE = Yb, Pr and Nd) single crystals, showing $(00l)$ ($l = \text{even integer}$) reflections. The insets are the corresponding optical photographs of RETi_3Bi_4 (RE = Yb, Pr and Nd) single crystals. (b) The enlarged (0010) peak of RETi_3Bi_4 (RE = Yb, Pr and Nd) single crystals. (c) The SAED pattern of YbTi_3Bi_4 along the

[0-26] zone axis. The inset is the thin specimen of YbTi_3Bi_4 etched by FIB. (d) The HAADF image collected along the [0-26] zone-axis, where the cyan, blue, and purple balls represent Yb, Ti, and Bi atoms, respectively.

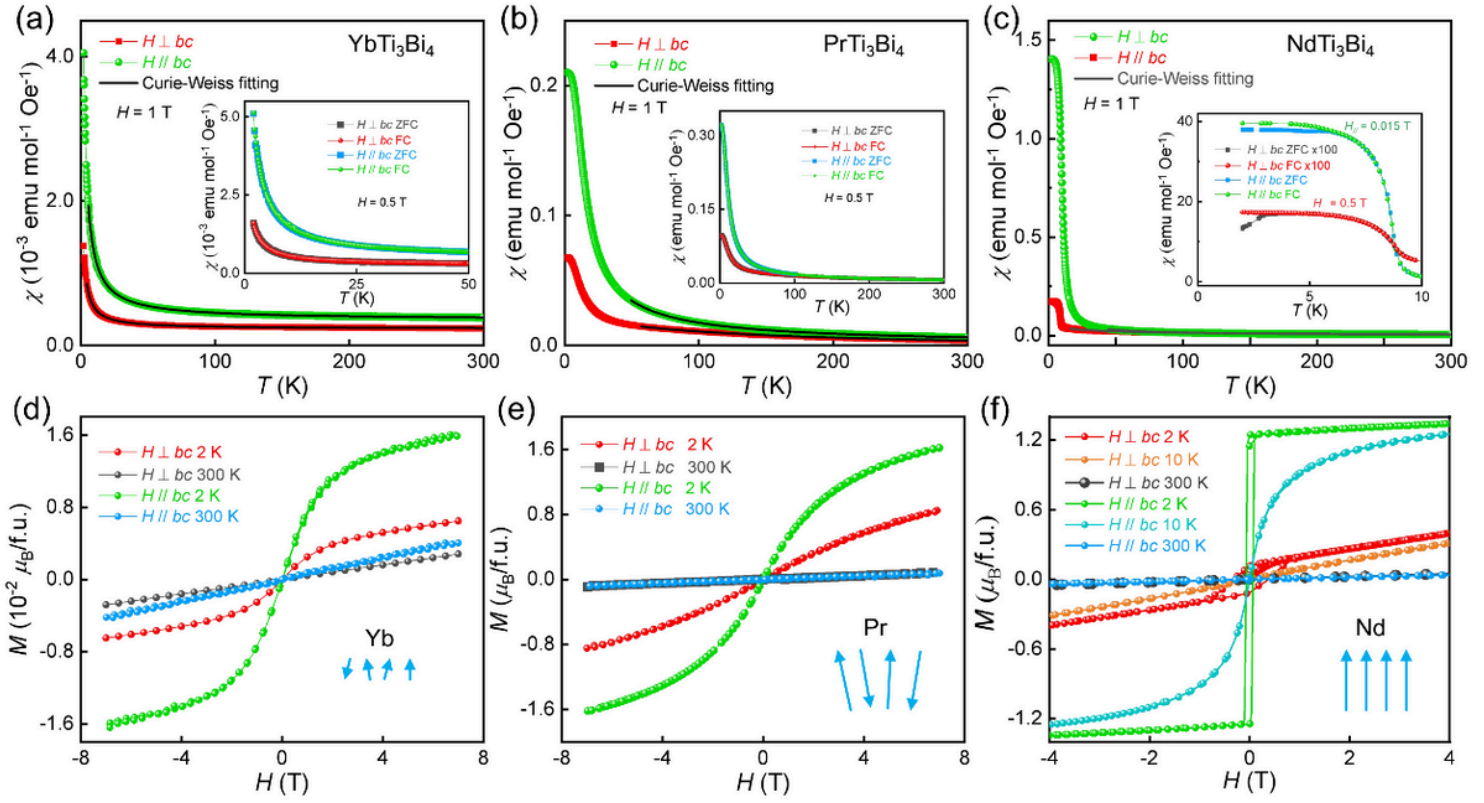


Figure 3

Various magnetic states of RETi_3Bi_4 (RE = Yb, Pr and Nd). Temperature-dependent magnetic susceptibility of (a) YbTi_3Bi_4 , (b) PrTi_3Bi_4 , and (c) NdTi_3Bi_4 single crystal with magnetic field (1 T) parallel and perpendicular to the bc plane. The black lines are Curie-Weiss fitting curves. The insets show the corresponding ZFC and FC curves under a smaller magnetic field. Field-dependent magnetization curves for (d) YbTi_3Bi_4 , (e) PrTi_3Bi_4 , and (f) NdTi_3Bi_4 single crystal with magnetic fields parallel and perpendicular to the bc plane. The insets show the schematic alignment of spin moments in the magnetic ground state of RETi_3Bi_4 .

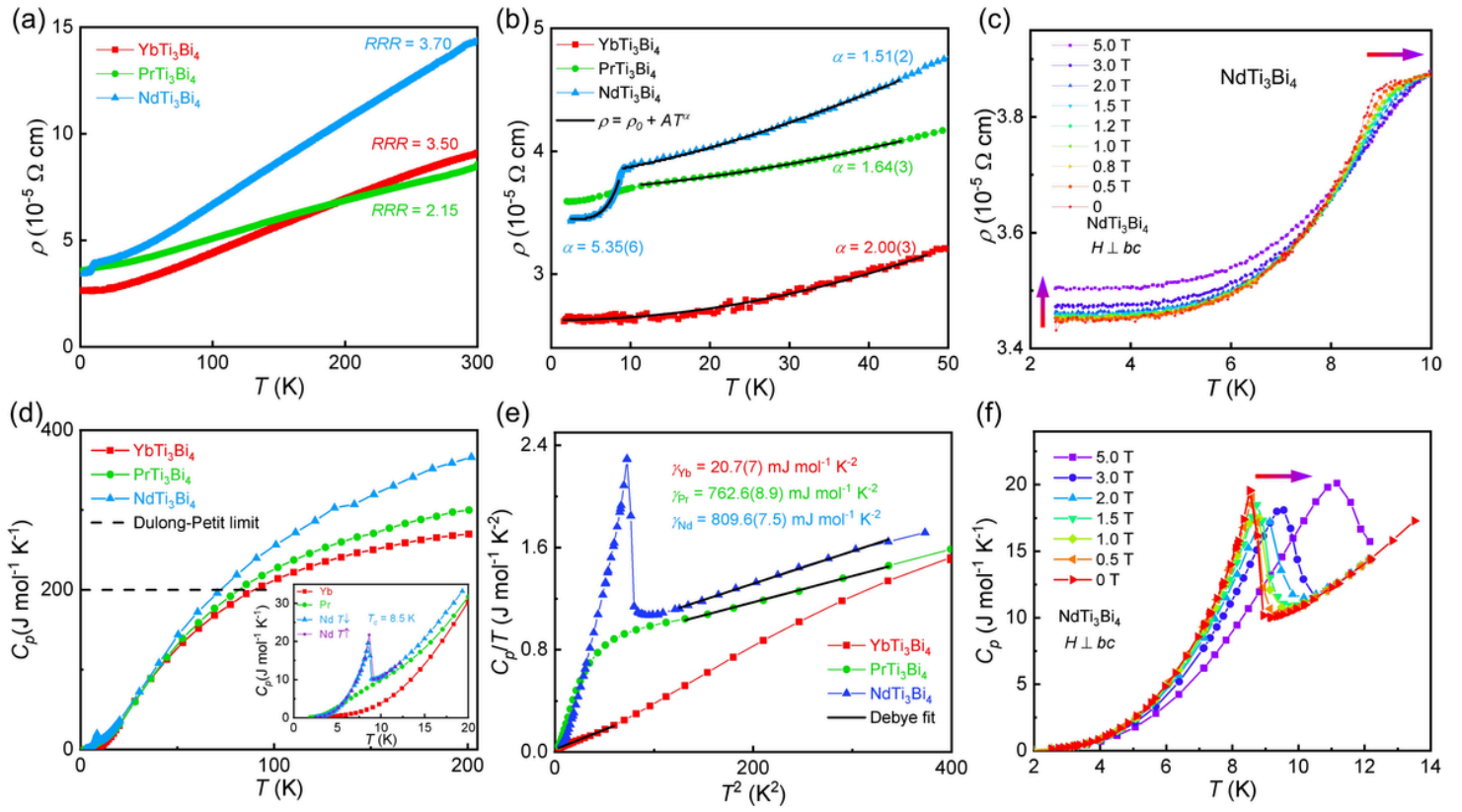


Figure 4

Electrical transport and specific heat capacity of RETi_3Bi_4 (RE = Yb, Pr and Nd). (a) Temperature-dependent in-plane resistivity (r_{bc}) of RETi_3Bi_4 (RE = Yb, Pr and Nd). (b) Power-law fittings of in-plane resistivity at low temperature. (c) In-plane resistivity of NdTi₃Bi₄ at low temperature under different magnetic field. (d) Temperature-dependent specific heat capacity of RETi_3Bi_4 (RE = Yb, Pr and Nd). The inset shows zoomed specific heat capacity at low temperature. The black dashed line shows the Dulong-Petit limit of RETi_3Bi_4 compounds. (e) C_p/T vs. T^2 plot for RETi_3Bi_4 (RE = Yb, Pr and Nd) and corresponding fittings (black lines) using the Debye model. (f) Zoomed specific heat capacity of NdTi₃Bi₄ at low temperature under different magnetic field.

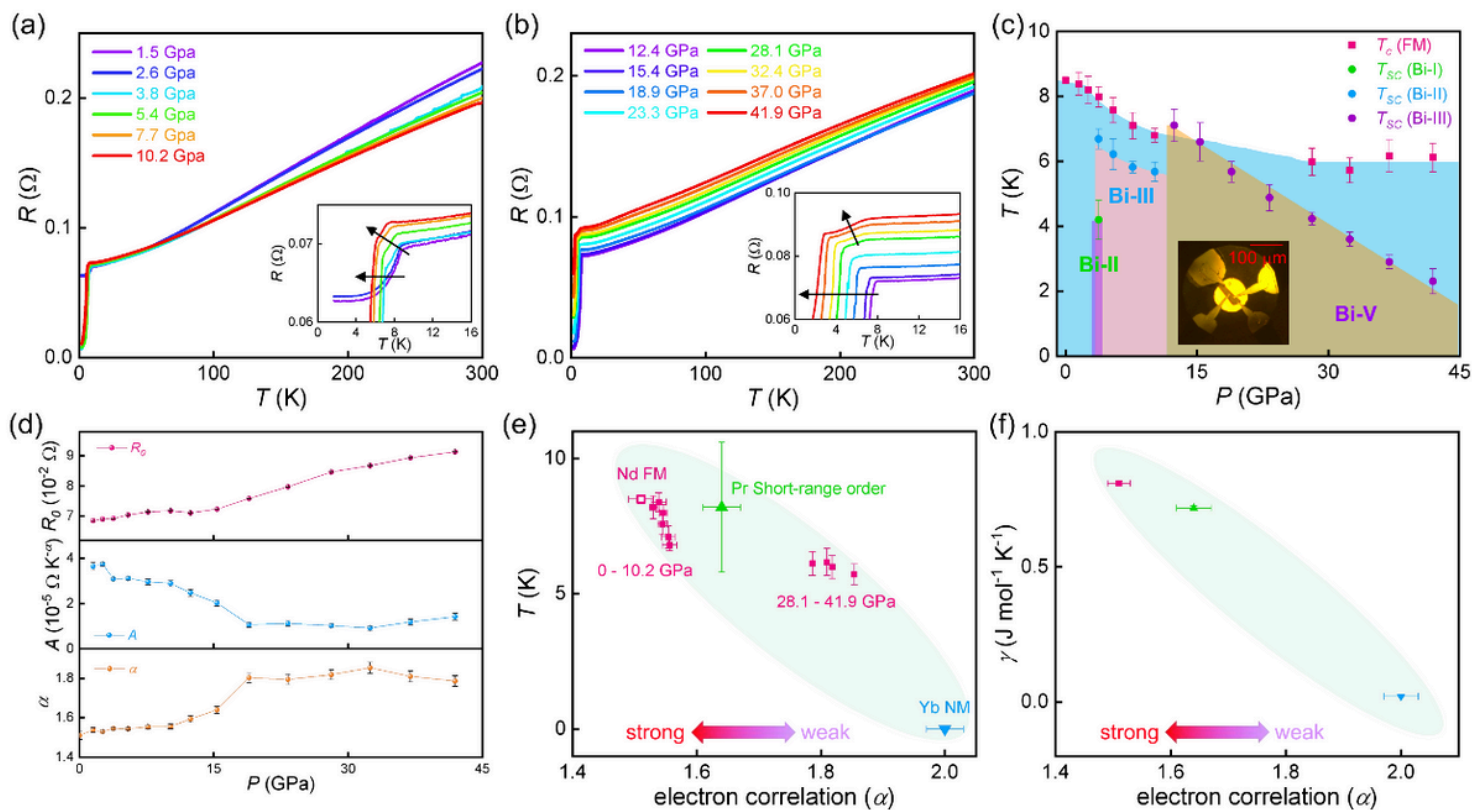


Figure 5

Magnetism and electron correlation tuned by pressure. Temperature-dependent resistance of NdTi_3Bi_4 under pressure from (a) 1.5 GPa to 10.2 GPa, and (b) 12.4 GPa to 41.9 GPa. The insets show the resistance at low temperature ranging from 1.7 K to 12 K. The arrows show the suppression trend of FM ordering temperature and superconducting transition temperature. (c) Pressure dependence of FM ordering temperature and superconducting transition temperature. The inset shows the optical photograph of four probe configuration in the diamond-anvil cell. (d) Fitted parameters of resistance at low temperature using Power law fitting $R = R_0 + AT^a$ under different pressure. (e) Evolution of FM ordering temperature with the value of power (a), which represents the strength of electron correlation. The corresponding ordering temperatures of YbTi_3Bi_4 and PrTi_3Bi_4 are taken to be zero and 8.2(2.0) K for comparison. (f) Evolution of Sommerfeld coefficient (γ) with the value of power (a).

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