Supporting information

Transition from ferromagnetic semiconductor to ferromagnetic

metal with enhanced Curie temperature in Cr₂Ge₂Te₆ via organic ion

intercalation

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S1. The discharge curve of (TBA)_xCr₂Ge₂Te₆

Figure S1 shows the typical discharge curve of $Cr_2Ge_2Te_6$ during the electrochemical intercalation process. For purpose of comparison, we set the blank control experiment, in which only $Cr_2Ge_2Te_6$ is not attached on the positive electrode but others remain the same.

For the blank control experiment, with applied current, the voltage quickly drops to -2V where electrolyte starts to decompose. In the electrochemical intercalation process of $(TBA)_x Cr_2 Ge_2 Te_6$, when x > 1.13, the voltage drops and keep at -2V, indicating that the electrolyte starts to decompose and no further intercalation would happen. Thus, from the discharge curve of $(TBA)_x Cr_2 Ge_2 Te_6$, the maximum *x*, namely the amount of intercalated TBA⁺ in $Cr_2 Ge_2 Te_6$, would be ~1.13.



Figure S1. The discharge curve of (TBA)Cr₂Ge₂Te₆ during the electrochemical intercalation process.

We also calculated the theoretical maximum amount of intercalated TBA⁺ in Cr₂Ge₂Te₆. As shown in figure S2, when TBA⁺ cations are "closely" packed, the packing density is calculated to be 0.35 nm² (= 1.19 × 1.19 /4, four tetrahedron would form a cubic) per TBA⁺ cation.^[1,2] Since in-plane area of a unit cell of the Cr₂Ge₂Te₆ is 0.4 (= 0.68 × 0.68 × $\sqrt{3}$ /2) and three slabs per unit cell, the theoretical maximum amount of *x* in (TBA)_{*x*}Cr₂Ge₂Te₆ is determined to be 1.14 (= 0.4 / 0.35 × 3 (three interlayer space) / 3 (Z=3)).



Figure S2. The "closely" packed structure model for the TBA⁺ intercalated phase.





Figure S3. (a) and (b): Temperature-dependent magnetic susceptibility of $Cr_2Ge_2Te_6$ for H // ab and H // c, respectively; **(c) and (d):** Field-dependent magnetic susceptibility of $Cr_2Ge_2Te_6$ with H // ab and H // c, respectively.

Figure S3 shows the temperature-dependent magnetic susceptibility and field-dependent

magnetic susceptibility of pristine $Cr_2Ge_2Te_6$ single crystals, in which the Curie temperature is 67 K. The saturation field is 5000 Oe for H // ab and 2500 Oe for H // c, respectively. Our data indicate that the magnetic easy-axis of $Cr_2Ge_2Te_6$ is <001> direction, which is in accordance with previous reports^[3,4].

S3. The fitting of energy gap in pristine Cr₂Ge₂Te₆ single crystal

We fitted the temperature-dependent resistivity with the thermal activation model, as described by the following formula:

$$\rho = \rho_0 \cdot exp(\frac{E_g}{2k_BT}),$$

where k_B is the Boltzmann constant, *T* is the temperature and E_g is the energy gap. From the linearly fitted curve (red line in Fig. S4(b)), E_g is determined to be ~ 0.22eV, which is in excellent agreement with previous reported value of ~ 0.2 eV^[4,5].



Figure S4. (a): Temperature dependent resistivity of $Cr_2Ge_2Te_6$; **(b):** Linear fit (red line) of the logarithmic plot of resistivity of $Cr_2Ge_2Te_6$ as a function of the reciprocal of temperature.

S4. The structure of TBA+

Figure S5 shows the schematic structure of tetrabutyl ammonium (TBA⁺) ion ^[6-9]. The lengths in different directions are labeled.



Figure S5. The schematic structure of tetrabutyl ammonium (TBA⁺).

S5. The HRTEM and SAED characterization of Cr₂Ge₂Te₆ and (TBA)Cr₂Ge₂Te₆



Figure S6. The cross-section HRTEM images of pristine $Cr_2Ge_2Te_6$ and hybrid superlattice (TBA) $Cr_2Ge_2Te_6$

Figure S6 shows the cross-section HRTEM images of $Cr_2Ge_2Te_6$ and hybrid superlattice (TBA) $Cr_2Ge_2Te_6$. In pristine $Cr_2Ge_2Te_6$, the interlayer spacing is 0.67 nm, which is in excellent agreement with the measurements from X-ray diffraction. After intercalation, the interlayer spacing increases to 1.59 nm in HRTEM images, and this value is slightly smaller than that of XRD measurements. This difference is likely due to a small angle deviation between the electron beam and the cross section plane of samples in the HRTEM measurements. Moreover, due to the relative low diffraction contrast, the intercalated TBA⁺ appears invisible in the HRTEM images. It is not noting that the layers in (TBA) $Cr_2Ge_2Te_6$ forms a "wavy" structure, which is also observed in organic molecule intercalated TiS₂^[10]. This is mainly because the organic molecules intercalated materials become much flexible compared with the mother compound.



Figure S7. The in-plane HRTEM and SAED images of (a) (TBA)Cr₂Ge₂Te₆; (b) Cr₂Ge₂Te₆.

Figure S7 shows the in-plane HRTEM and SAED images of intercalated (TBA) $Cr_2Ge_2Te_6$ and pristine $Cr_2Ge_2Te_6$. For intercalated (TBA) $Cr_2Ge_2Te_6$, most of the area observed in TEM is within the in-plane direction. From the HRTEM and SAED result, we find that the in-plane lattice parameter of intercalated (TBA) $Cr_2Ge_2Te_6$ remains almost the same with pristine $Cr_2Ge_2Te_6$. This result is in accordance with other organic intercalated compounds such as TaS₂, TiS₂ and MoO₃ ^[11-13]. Moreover, the (200) reflection appears in (TBA) $Cr_2Ge_2Te_6$, which could not be observed in pristine $Cr_2Ge_2Te_6$ due to extinction. This result suggests that the symmetry of (TBA) $Cr_2Ge_2Te_6$ is reduced compared with $Cr_2Ge_2Te_6$.



Figure S8. The TEM EDS result of $(TBA)Cr_2Ge_2Te_6$ and $Cr_2Ge_2Te_6$. Three average spectra shows that Cr:Ge:Te = 1 : 0.99 : 3.01 in $(TBA)Cr_2Ge_2Te_6$ and Cr:Ge:Te = 1 : 1.01 : 2.98 in $Cr_2Ge_2Te_6$.

Figure S8 shows the TEM Energy Dispersive Spectroscopy (EDS) on (TBA) $Cr_2Ge_2Te_6$ and $Cr_2Ge_2Te_6$. Three average spectra shows that Cr:Ge:Te = 1 : 0.99 : 3.01 in (TBA) $Cr_2Ge_2Te_6$ and Cr:Ge:Te = 1 : 1.01 : 2.98 in $Cr_2Ge_2Te_6$, respectively. It is worth noting that a little amount of N could be detected in (TBA) $Cr_2Ge_2Te_6$, which confirms that the observed area shown in Figure S7(a) is (TBA) $Cr_2Ge_2Te_6$.



S6. The Curie-Weiss fitting of the magnetic susceptibility of (TBA)Cr₂Ge₂Te₆

Figure S9. The inversed temperature-dependent susceptibility of (TBA) $Cr_2Ge_2Te_6$. The red solid line shows the Curie-Weiss fitting of the susceptibility curves. T^{mag}_{C} is the ferromagnetic order temperature of (TBA) $Cr_2Ge_2Te_6$, which is labeled as the vertical red line.

Figure S9 shows the inversed temperature-dependent susceptibility of (TBA)Cr₂Ge₂Te₆ with the magnetic field applied in different directions. By fitting the data with Curie-Weiss law, we obtained effective magnetic moment $\mu_{eff} \sim 4.20 \ \mu_{B} / \text{Cr} (H // \text{ab})$ and $4.18 \ \mu_{B} / \text{Cr} (H // \text{c})$.

S7. Elemental analysis and thermo-gravimetric analysis (TGA)

In order to accurately measure the amount of intercalated TBA⁺, we conduct elemental analysis with CHN analyzer. The absolute C, H, N content in the intercalated (TBA)Cr₂Ge₂Te₆ sample is determined to be 15.13 wt.%, 3.14 wt.% and 1.14 wt.%, which is consistent with the C, H, N content of 15.28 wt.%, 2.88 wt.% and 1.11 wt.% from the chemical formula (TBA)Cr₂Ge₂Te₆. Moreover, the EDS result presented in Figure S8 has confirmed that Cr:Ge:Te

 \approx 1:1:3 in (TBA)Cr₂Ge₂Te₆ and Cr₂Ge₂Te₆.

Figure S10 shows the thermo-gravimetric analysis (TGA) for both (TBA) $Cr_2Ge_2Te_6$ and $Cr_2Ge_2Te_6$. The temperature range is 30-400 °C and the whole experiment is conducted under Nitrogen gas. For pristine $Cr_2Ge_2Te_6$, there is no obvious weight loss in the measured temperature range. However, for intercalated (TBA) $Cr_2Ge_2Te_6$, there is obvious weight loss during the heating process and finally reaches a platform at 400 °C with weight loss of 18.1 %. This weight loss just coincides with the weight ratio of TBA⁺ in (TBA) $Cr_2Ge_2Te_6$, which confirms the composition of the intercalated compounds. Thus, combination of these different chemical analysis indicates that the composition of the intercalated compound is (TBA) $Cr_2Ge_2Te_6$.



Figure S10. The thermo-gravimetric analysis (TGA) for both (TBA)Cr₂Ge₂Te₆ and Cr₂Ge₂Te₆

S8. Annealing experiment of (TBA)Cr₂Ge₂Te₆

We carried out the annealing experiment of (TBA)Cr₂Ge₂Te₆ at different temperature. Figure S11 shows the XRD, magnetic susceptibility and resistivity measurement results of original (TBA)Cr₂Ge₂Te₆ sample and annealed sample (150 °C and 300 °C). To perform the experiment, one single crystal (TBA)Cr₂Ge₂Te₆ is cut into two pieces and sealed separately into two quartz tubes under vacuum (< 10⁻³ Pa). The two pieces of sample are annealed at 150 °C and 300 °C for 2 hours, respectively.

For the original (TBA) $Cr_2Ge_2Te_6$ sample, the XRD, magnetic susceptibility and resistance results are in accordance with that illustrated in manuscript. After annealed at 150 °C under vacuum for 2 hours, the ferromagnetic transition temperature (T_c) of (TBA) $Cr_2Ge_2Te_6$ drops to 65 K and the temperature dependent resistance shows insulating behavior, being consistent with $Cr_2Ge_2Te_6$. Moreover, the original XRD peaks vanish and new peaks located at

 26.00° and 53.42° emerge, corresponding to the (006) and (00 12) peaks of $Cr_2Ge_2Te_6$. Combined the annealing experiment with TGA experiment, we can conclude that the intercalated organic TBA⁺ cations could be easily de-intercalated with annealing at low temperature, which could be ascribed to the greatly enlarged interlayer spacing and very weak interlayer interaction. More importantly, it unambiguously proves that the large increase of Curie temperature is caused by the intercalation of TBA⁺ cations in (TBA) $Cr_2Ge_2Te_6$.



Figure S11. (a) The XRD patterns; (b) The temperature dependent resistance; (c) The temperature dependent magnetic susceptibility of the original (TBA)Cr₂Ge₂Te₆ and annealed sample.

S9. The heat capacity of Cr₂Ge₂Te₆ and (TBA)Cr₂Ge₂Te₆

Figure S12 shows the heat capacity of $Cr_2Ge_2Te_6$ and $(TBA)Cr_2Ge_2Te_6$ as a function of temperature. For pristine $Cr_2Ge_2Te_6$, a sharp peak appears at T = 65 K, indicating the magnetic transition temperature, which is consistent with magnetic susceptibility measurements. For (TBA)Cr_2Ge_2Te_6, the characteristic broad peak shows up at T = 206 K, being consistent with

the ferromagnetic phase transition temperature as revealed by the magnetic susceptibility and resistance measurement. The heat capacity of hybrid superlattice (TBA)Cr₂Ge₂Te₆ is much larger than that of pristine Cr₂Ge₂Te₆, which could be attributed to the intercalation of organic TBA⁺ cations, given that the heat capacity of TBAB (Tetrabutyl ammonium bromide) is significant and has a value of 448 J mol⁻¹ K⁻¹ at 300 K^[14].



Figure S12. The heat capacity of Cr₂Ge₂Te₆ and (TBA)Cr₂Ge₂Te₆

S10. The details on the DFT calculation

At initial, we intend to do the calculation with simulating the atomic position of TBA⁺. However, simulating realistic compound with intercalated TBA⁺ cations (> 150 atoms) is far beyond our computation ability. Moreover, in principle, TBA⁺ cation does not strongly interact with $Cr_2Ge_2Te_6$ -slab and act as an inert charged object with Fermi-levels far away below the Fermi energy. Thus, based on the assumption that intercalated TBA⁺ cation only enlarges the vacuum distance and $Cr_2Ge_2Te_6$ -slab is electron doped, we adopt the following model and parameters:

(1). According to the previous mentioned structure characterization result, we adopt experimental hexagonal unit cell with lattice constants (a = b = 6.83 Å) and atomic positions.^[15] The lattice constant c is expanded to experimental value c = 49.44 Å, with the thickness ~ 3.308 Å of each $Cr_2Ge_2Te_6$ unit and the inter-slab separation to 13.172 Å to accommodate the organic cation (TBA⁺) (The inter-slab separation is defined as the vacuum distance between Te atoms of the nearest neighbor $Cr_2Ge_2Te_6$ -slab). The unit cell of $Cr_2Ge_2Te_6$ and (TBA) $Cr_2Ge_2Te_6$ is shown in figure S13.

(2). Furthermore, the charges +3e per unit cell, which act for the electrons migrated from Ag^+ to the $Cr_2Ge_2Te_6$ -slab, are taken into account using the charged cell method.

(3). We perform the GGA + U + SO calculations with U = 0.5 for bulk $Cr_2Ge_2Te_6$ according to literature.^[16] After the intercalation, the ground state of the compound transits from insulator to metal, suggesting that the effective Coulomb interaction U should be obviously reduced due to the screening effects among conducting electrons. Thus, we adopt U = 0.2 eV for the calculation of intercalated (TBA)Cr₂Ge₂Te₆.



Figure S13. The unit cell of $Cr_2Ge_2Te_6$ and $(TBA)Cr_2Ge_2Te_6$. The TBA cation is included yet not considered in calculation.



S11. Calculated band structure of Cr₂Ge₂Te₆ and (TBA)Cr₂Ge₂Te₆

Figure S14. The calculated band structure of Cr₂Ge₂Te₆ and (TBA)Cr₂Ge₂Te₆.

S12. Spin-polarized full DOS of Cr₂Ge₂Te₆ and (TBA)Cr₂Ge₂Te₆



Figure S15. The calculated spin-polarized full DOS of Cr₂Ge₂Te₆ and (TBA)Cr₂Ge₂Te₆.

S13. Raman spectroscopy, X-ray photoelectron spectroscopy (XPS) and Fourier transform infrared spectroscopy (FT-IR) of Cr₂Ge₂Te₆ and (TBA)Cr₂Ge₂Te₆

Figure S16 shows the Raman spectroscopy of intercalated (TBA)Cr₂Ge₂Te₆, pristine Cr₂Ge₂Te₆ as well as TBAB (tetrabutyl ammonium bromide). The Raman measurements are performed at ambient temperature on a MonoVista CRS+ instrument with 532-nm line as the excitation source. All the samples are sealed in a stage under inert gas. For pristine Cr₂Ge₂Te₆, five modes, positioned at 110.5 cm⁻¹(E_g²), 135.6 cm⁻¹(A_g¹), 211.8 cm⁻¹(E_g⁴), 233.4 cm⁻¹(E_g⁵), and 292.9 cm⁻¹(A_g²), could be observed and agree well with the previous report.^[17] After intercalation, the five modes still exists but shifted to 108.8 cm⁻¹(E_g²), 130.6 cm⁻¹(A_g¹), 211.2 cm⁻¹(E_g⁴), 231.8 cm⁻¹(E_g⁵), and 291.7 cm⁻¹(A_g²). Besides, there appears a new mode at 259.8 cm⁻¹, which is in accordance with the Raman mode of TBAB and proves the intercalation of TBA⁺ in (TBA)Cr₂Ge₂Te₆. Compared with Cr₂Ge₂Te₆, it is apparently that all Raman modes move to lower wavenumber, which suggests the electron doping in (TBA)Cr₂Ge₂Te₆. Especially, the A_g¹

mode moves the most, with ~ 5 cm⁻¹. From the analysis for the Raman mode of $Cr_2Ge_2Te_6$, it can be seen that A_g^{-1} is the out-of-plane phonon mode.^[17] With the intercalation of TBA⁺ cations, the interlayer interaction reduces a lot. Thus, the reduced interlayer interaction as well as electron doping lead to the dramatic change of A_g^{-1} mode in (TBA) $Cr_2Ge_2Te_6$. The Raman spectroscopy experiment not only proves the that the intercalated (TBA) $Cr_2Ge_2Te_6$ owns similar crystal structure with pristine $Cr_2Ge_2Te_6$, but also confirms the electron doping in (TBA) $Cr_2Ge_2Te_6$.

Figure S17 shows the X-ray photoelectron spectroscopy (XPS) of $Cr_2Ge_2Te_6$ and (TBA) $Cr_2Ge_2Te_6$ with the region of Cr 2p. It can be seen that the Cr $2p_{1/2}$ and Cr $2p_{3/2}$ peaks of (TBA) $Cr_2Ge_2Te_6$ shifted to lower binding energy compared with pristine $Cr_2Ge_2Te_6$, which proves the reduction of Cr as well as the electron doping scenario after intercalation.

Figure S18 shows the FT-IR spectroscopy of $Cr_2Ge_2Te_6$, TBAB and (TBA) $Cr_2Ge_2Te_6$. There is no obvious absorption band for $Cr_2Ge_2Te_6$ at the range of 500-4000 wave number (cm⁻¹). However, the FT-IR spectroscopy of (TBA) $Cr_2Ge_2Te_6$ resembles some feature peaks like that of TBAB, indicating the successful intercalation of TBA⁺ cation into $Cr_2Ge_2Te_6$.



Figure S16. The Raman spectroscopy of TBAB, Cr₂Ge₂Te₆ and (TBA)Cr₂Ge₂Te₆.



Figure S17. The X-ray photoelectron spectroscopy (XPS) of $Cr_2Ge_2Te_6$ and $(TBA)Cr_2Ge_2Te_6$ with the region of Cr 2p.



Figure S18. The FT-IR spectroscopy of TBAB, Cr₂Ge₂Te₆ and (TBA)Cr₂Ge₂Te₆.

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