Supplement materials:

Understanding CrGeTe₃: an abnormal phase change material with inverse resistance and density contrast

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Fig. S1 a) Pair distribution functions of Te-Ge, Te-Te and Ge-Ge. b) Proportion of Cr-

Cr, Cr-Ge and Cr-Te bonds.



Fig. S2 Coordination numbers of Ge (a) and Te (b) in amorphous CrGeTe₃.



Fig. S3 Bader charge of CrGeTe₃ crystal.



Fig. S4 Average CHOPs of Cr-Cr, Cr-Ge and Cr-Te bonds. None of them have clear antibonding interactions below the fermi level, thus all the Cr bonds are stable. Besides, these three kinds of bonds have almost the same bonding contributions, showing approximate bonding strength.



Fig. S5 DFT+U and spin-polarization methods are used to calculate energy band structure of $CrGeTe_3 crystal$. The band gap is about 0.31eV.



Fig. S6 Atomic plane (100) corresponding to the charge density and ELF in Fig. 6 (d, e).



Fig. S7 a) The 3D display of 0.78 ELF isosurface of c-CrGT. The "yellow hat" areas show the place of lone pair electrons. b-c) The ELF and atoms distribution of (100) plane of c-CrGT. The "red hat" regions show the highly localized lone pair. It confirms that ELF is a valid method to identify lone-pair electrons.



Fig. S8 The ELF and atomic distribution of (110) plane of a-CrGT. It shows many

lone-pair electrons in amorphous CrGeTe₃.



Fig. S9 a) Amorphous a-CrGT model includes 180 atoms. b) Amorphous a-CrGT model includes 270 atoms. c) Amorphous a-CrGT model includes 180 atoms in irregular box. We have built a series of amorphous models of various sizes, original structure and quenching process. All of them show smaller mass density than crystal and obvious tight Cr clusters. And we also observe the distribution of long and short Cr-Cr bond lengths in these models.