

Electronic Properties and Phase Transition in the Kagome Metal $\text{Yb}_{0.5}\text{Co}_3\text{Ge}_3$

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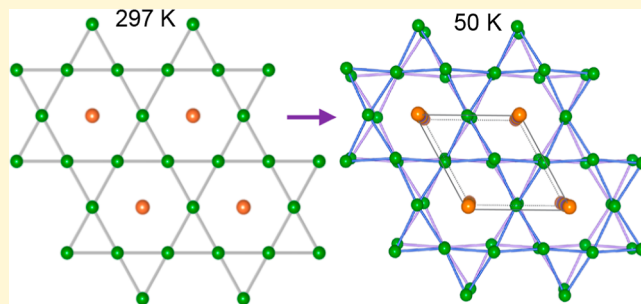


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Supporting Information

ABSTRACT: The Kagome lattice is an important fundamental structure in condensed matter physics for investigating the interplay of electron correlation, topology, and frustrated magnetism. Recent work on Kagome metals in the AV_3Sb_5 ($A = \text{K}, \text{Rb}$, and Cs) family has shown a multitude of correlation-driven distortions, including symmetry breaking charge density waves and nematic superconductivity at low temperatures. Here, we study the new Kagome metal $\text{Yb}_{0.5}\text{Co}_3\text{Ge}_3$ and find a temperature-dependent kink in the resistivity that is highly similar to the AV_3Sb_5 behavior and is commensurate with an in-plane structural distortion of the Co Kagome lattice along with a doubling of the c -axis. The symmetry is lower below the transition temperature, with a breaking the in-plane mirror planes and C_6 rotation, while gaining a screw axis along the c -direction. At very low temperatures, anisotropic negative magnetoresistance is observed, which may be related to anisotropic magnetism. This raises questions about the types of the distortions in Kagome nets and their resulting physical properties including superconductivity and magnetism.



INTRODUCTION

Materials containing 2D Kagome lattices, also known as trihexagonal tiling, are of extreme interest in condensed matter physics today. The Kagome lattice is made up of hexagons that are surrounded on their edges by equilateral triangles. Analogous to the honeycomb lattice, the Kagome lattice is an important structure for realizing a quantum spin liquid state due to its inherent geometric frustration; some of the leading spin liquid candidates are Kagome insulators.^{1,2} Recently, however, Kagome metals have gained attention due to the realization of both topological Dirac electrons and flat bands and van Hove singularities resulting in strong electron correlation.^{3–7} Materials in the AV_3Sb_5 ($A = \text{K}, \text{Cs}$, and Rb) family^{8–11} have shown both topological bands with low effective mass, a large anomalous Hall effect,^{12,13} and a cascade of charge density wave (CDW)^{14–23} orderings and superconductivity^{24–30} as temperature is lowered.

The intertwining of orders seen in Kagome metals is a complicated interplay of electronic correlation, topology, and magnetism, resulting in nearby ground states that are accessible by temperature control. While some phases (AV_3Sb_5) have shown electronic correlation-related charge orders and topology, others have shown topology and magnetism.^{31–39} Studies are ongoing on Kagome systems which show correlation and magnetism, where charge/spin orders intertwine with magnetic orders of a spin sublattice. It has been shown that the structural distortions of the Kagome lattice and

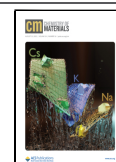
symmetry breaking are closely related to the appearance of new orders, including the symmetry breaking CDW order observed in AV_3Sb_5 compounds and antiferromagnetic FeGe .⁴⁰ How the various orders work together, or compete with each other, and what effect that has on structural distortions in the Kagome net and the resulting physical properties remain an active area of investigation.

In this paper, we report the presence of a kink in the resistivity at 95 K, below which there is a structural phase transition resulting in the distortion of the Co Kagome net, in the new Kagome metal $\text{Yb}_{0.5}\text{Co}_3\text{Ge}_3$. Through single-crystal X-ray diffraction structure determination, the low-temperature structure was found to break C_6 rotation symmetry and out-of-plane mirror planes, lowering symmetry from the $P6/mmm$ space group to $P6_3/m$. In addition, a small upturn is evident in the resistivity below 18 K, commensurate with a previously seen transition in magnetization. Below this temperature, anisotropic negative magnetoresistance is observed, in contrast with the positive magnetoresistance seen in other Kagome metals such as AV_3Sb_5 . $\text{Yb}_{0.5}\text{Co}_3\text{Ge}_3$ is a good platform for

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studying the effect of correlation and magnetism on the electronic properties in a Kagome system and adds to the ongoing effort to understand the phase space of Kagome metals.

EXPERIMENTAL SECTION

Synthesis and Property Measurements. Single crystals of $\text{Yb}_{0.5}\text{Co}_3\text{Ge}_3$ were grown as reported previously.⁴¹ Elements were weighed out into a Canfield crucible set⁴² with a molar ratio of 3:2:7:52 Yb/Co/Ge/Sn, respectively, and sealed in a fused silica tube filled with argon gas $\sim 1/3$ atm pressure. The sealed ampoule was then heated in a furnace to 1175 °C at 100 °C/h and dwelled for 24 h before cooling the ampoule down to 815 °C at a rate of 3 °C/h. The ampoule was then removed from the furnace, inverted, and centrifuged to remove the excess flux from the crystals. The needle-shaped crystals were then etched with dilute HCl followed by dilute HNO_3 etching to remove residual flux from the crystal surface. Silver paste was used to make contact with $\text{Yb}_{0.5}\text{Co}_3\text{Ge}_3$ crystals, and the electrical properties were measured in a Quantum Design physical property measurement system (PPMS) using the four-probe method with an a.c. current applied along the c -axis.

Single-Crystal X-ray Diffraction. For comparison, the crystal structure of $\text{Yb}_{0.5}\text{Co}_3\text{Ge}_3$ collected at room temperature⁴¹ is compared to the low-temperature model. The 50 K single-crystal X-ray diffraction data were collected using a Rigaku XtaLAB AFC12-(RCD3) diffractometer equipped with graphite monochromated Mo $K\alpha$ radiation ($\lambda = 0.71073$ Å), HyPix-6000He area detector, and Rigaku Oxford Diffraction CrysAlisPro software. Sample temperature was controlled with the dual flow nitrogen and helium gas cooler N-Helix by Oxford Cryosystems. The numerical absorption correction was completed based on Gaussian integration over a multifaceted crystal model. The empirical absorption correction was carried out using spherical harmonics implemented in the SCALE3 ABSPACK scaling algorithm in CrysAlis PRO 1.171.41.123a (Rigaku Oxford diffraction 2022). The 50 K data were modeled in Jana 2006 software⁴³ using the room temperature structure as the preliminary structural model. Table 1 provides the unit cell parameters and space group at room temperature and 50 K.

Table 1. Unit Cell Parameters from Single-Crystal X-ray Diffraction

	297 K	50 K
formula	$\text{Yb}_{0.5}\text{Co}_3\text{Ge}_3$	$\text{Yb}_{0.5}\text{Co}_3\text{Ge}_3$
space group	$P6/mmm$	$P6_3/m$
lattice parameters		
a (Å)	5.0949(10)	5.0705(4)
c (Å)	3.9136(9)	7.7780(9)
V (Å ³)	87.98(4)	173.18(3)
Z	1	2

RESULTS AND DISCUSSION

$\text{Yb}_{0.5}\text{Co}_3\text{Ge}_3$ crystallizes in the hexagonal $P6/mmm$ space group at room temperature⁴¹ and adopts a hybrid structure of the YCo_6Ge_6 and CoSn prototypes^{44–46} with a Co Kagome lattice; the crystal structure is shown in Figure 1a. The comparison between these structures is also included in the Supporting Information.

Single crystals of $\text{Yb}_{0.5}\text{Co}_3\text{Ge}_3$ present a rod-like shape with the long axis being the c -axis.⁴¹ Figure 1b shows the temperature-dependent resistivity $\rho(T)$ curve, which presents metallic behavior with reducing temperature. Several points of interest are observed on the $\rho(T)$ curve; the first one is an evident kink near $T^* \sim 95$ K with a clear transition on the $d\rho(T)/dT$ versus T curve (lower inset Figure 1b). This is

found to correspond to a structural phase transition, which will be discussed in detail below. Below ~ 18 K, a weak upturn of the resistivity is observed (also shown by the change in sign of the $d\rho(T)/dT$ versus T curve in the inset), followed by a superconductor-like transition ~ 3.6 K (upper inset Figure 1b). Above the upturn of the resistivity, the low-temperature $\rho(T)$ data (23–60 K) are well fitted using the equation $\rho(T) = \rho(0) + AT^n$ (black line in Figure 1b), with $n = 2.04$, $\rho(0) \approx 14.7 \mu\Omega \text{ cm}$, and $A \approx 0.00127 \mu\Omega \text{ cm K}^{-2}$, suggesting that electron–electron interactions dominate electronic transport in this regime.^{34,47}

The magnetoresistance is measured to study the magnetic response at low temperature. Figure 1c shows the $\rho(H)$ data measured with magnetic field applied in the Kagome plane ($H \perp I$) and perpendicular to the Kagome plane ($H // I$) at 2 K. The rapid increase in $\rho(H)$ at a very small field ($\mu_0 H_c \sim 15$ mT for $H \perp I$ and $\mu_0 H_c \sim 20$ mT for $H // I$) confirms the superconducting transition. Since the resistivity does not reach zero (only an $\sim 30\%$ drop) and the superconducting signal is very close to the superconductivity of Sn ($T_c \sim 3.72$ K and $\mu_0 H_c \sim 30$ mT), it is likely that the superconductivity is not intrinsic but is rather coming from a Sn flux residual, although the sample was carefully centrifuged after growth and etched in dilute HCl and HNO_3 to remove the residual of Sn flux. Investigation with doping and extremely low temperature study are necessary to search for superconductivity in this material, which is beyond the scope of this work.

After breaking superconductivity, a negative magnetoresistance is observed to high field, which is stronger for the magnetic field applied along the c -axis ($H // I$) compared with applying the magnetic field in the Kagome plane ($H \perp I$). Figure 1d shows the temperature dependence of magnetoresistance measured with $H \perp I$, indicating that the negative magnetoresistance appears in the upturn region of resistivity (see Figure S2 for ρ vs T data in the Supporting Information). Kagome metals with weak magnetism typically present positive magnetoresistance, such as the AV_3Sb_5 family,^{12,13} and some CoSn -type materials.⁴⁸ Negative magnetoresistance has been seen in ferromagnetic Kagome materials and was also proposed to occur from electron correlation-induced ferromagnetic spin fluctuations.^{48,49} For $\text{Yb}_{0.5}\text{Co}_3\text{Ge}_3$, an earlier study on the magnetic behavior indicated that the compound has antiferromagnetic coupling without a clear long-range magnetic order.⁴¹ In that study, below 15–20 K, a weak transition of magnetization was reported which was proposed to arise from spin canting or spin reorientation, and a larger magnetization was reported for the out-of-plane applied magnetic field (along the c -axis) compared with in the Kagome plane.⁴¹ The observed upturn of resistivity below 18–25 K in this work aligns very well with the temperature of the previously reported magnetization transition. These results indicate that the appearance of negative magnetoresistance is related to the magnetization transition, and the anisotropic negative magnetoresistance shown in Figure 1c may arise from the anisotropic magnetism in $\text{Yb}_{0.5}\text{Co}_3\text{Ge}_3$ at low temperature. Detailed study on the magnetic behavior of the phase in the future may reveal the origin of the transition and the negative magnetoresistance.

The main result of this work is the prominent transition near 95 K in the $\rho(T)$. Very similar transition features were observed in the $\rho(T)$ of some Kagome metals, such as the AV_3Sb_5 ($A = \text{K}, \text{Cs}, \text{and Rb}$) family⁸ and the antiferromagnetic Kagome metal FeGe ,⁴⁰ between 75 and 100 K. These

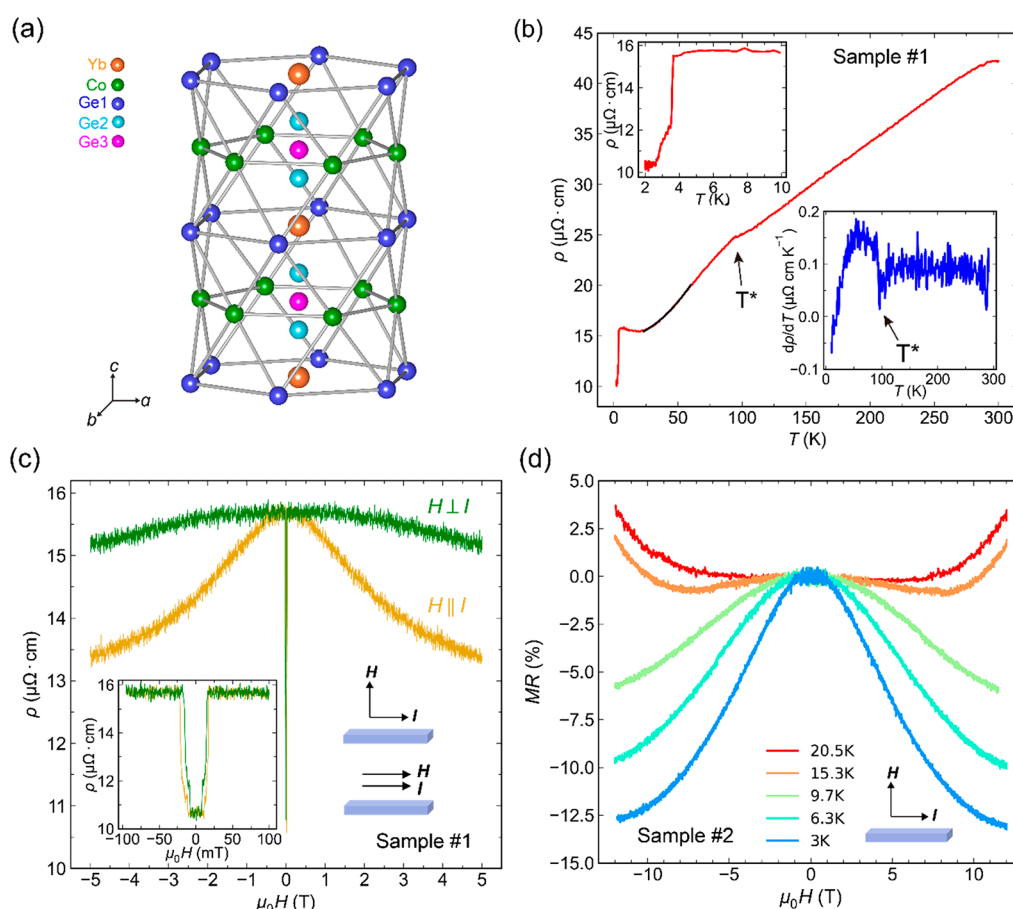


Figure 1. Structure and properties of $\text{Yb}_{0.5}\text{Co}_3\text{Ge}_3$. (a). Crystal structure of $\text{Yb}_{0.5}\text{Co}_3\text{Ge}_3$ in the $P6_3/mmm$ phase. (b). Temperature dependence of the resistivity with current applied along the c -axis. The top inset is the ρ vs T curve at low temperature, and the bottom inset is the $d\rho/dT$ vs T curve. The transition near 95 K is marked by a black arrow. The black line on the ρ vs T curve in the main panel is a fit to $\rho(T) = \rho(0) + AT^n$. (c). Magnetic field dependence of the resistivity measured at 2 K. The inset on the left is the zoom-in of the superconductivity at a small field. A schematic of the applied magnetic field direction for $H \parallel I$ and $H \perp I$ is shown in the inset on the right. (d). Temperature dependence of magnetoresistance measured, and resistivity is shown in S12 for $H \perp I$.

transitions were found to be the result of superlattice formation from CDW ordering.^{15–18,40} To reveal whether there was a corresponding structural transition in $\text{Yb}_{0.5}\text{Co}_3\text{Ge}_3$, we performed single-crystal X-ray diffraction above and below the transition temperature. Figure 2a,b shows the projection of the X-ray diffraction pattern along the a -axis (b – c plane) and c -axis (a – b plane) in reciprocal space, respectively. In the $(0kl)$ plane (b – c plane), a new set of diffraction peaks (marked by the orange box) can be seen in the 50 K pattern, located at half-integer spacing along the l direction (c -axis in real space, Figure 2a), compared with the diffraction pattern measured at room temperature. These new peaks arise from the formation of a superlattice with a doubled unit cell along the c -axis. In the (hko) plane (a – b plane), a new set of diffraction peaks (marked by the yellow circles) is also observed at 50 K compared with the 297 K pattern (Figure 2b). These peaks do not arise from a superlattice formation in the a – b plane but instead become visible at 50 K due to distortion of the Kagome lattice.

The lattice parameters of $\text{Yb}_{0.5}\text{Co}_3\text{Ge}_3$ obtained at 297 K and at 50 K from the single-crystal X-ray diffraction are provided in Table 1, and the corresponding crystal structure is shown in Figure 3. The crystal structure of $\text{Yb}_{0.5}\text{Co}_3\text{Ge}_3$ can be described as a stacking of alternating subunits—a subunit with a hexagonal array (or honeycomb arrangement) of Ge atoms

that can be partially stuffed in-plane with Yb and the transition metal (Co) Kagome net that contains Ge atoms within the hexagons which can be displaced out-of-plane in response to partial occupation of the Yb above or below them (Figure 3a,b). The spacing between the planes of these subunits along the c -axis is ~ 1.96 Å at room temperature and ~ 1.94 Å at 50 K, a relatively insignificant contraction along the c -axis. At room temperature, the tiling of hexagons and triangles in the Kagome net adopts ideal internal bond angles of 120° and 60° , respectively, with both types of geometric arrangements containing a uniform Co–Co interatomic distance of ~ 2.55 Å, being a perfect Kagome net, as shown in Figure 3c.

At low temperature, the crystal structure of $\text{Yb}_{0.5}\text{Co}_3\text{Ge}_3$ is best modeled in the $P6_3/m$ space group due to a geometric distortion within the plane of the Kagome net. This is not driven by either the Yb or Ge atoms, which retain their geometric relations very closely with the room temperature structure, but rather from the deviation of the Co atoms from the ideal positions of the Kagome lattice. The triangular arrangements remain unaltered (or temperature-independent) in their interatomic distances but are slightly rotated relative to each other (Figure 3d), resulting in the hexagonal arrangements becoming distorted. The bond angles in the hexagons therefore deviate by $\pm 7.5^\circ$ from the ideal internal bond angles of 120° in an alternating fashion when comparing angles

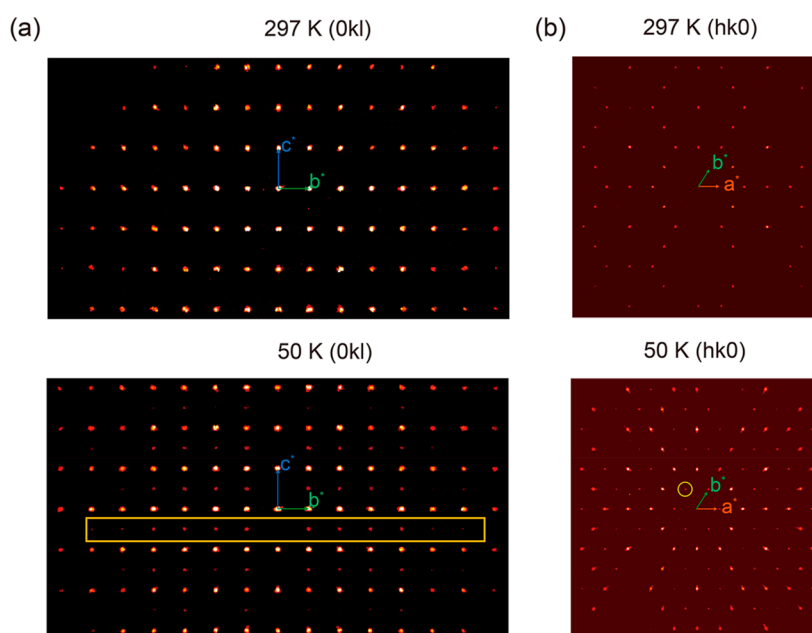


Figure 2. X-ray diffraction pattern. (a) Projection of the diffraction pattern along the a -axis ($0kl$ plane) at 297 and 50 K. The appearance of superlattice peaks (marked by the orange rectangle) at 50 K is due to the doubling of the c -axis. (b) Projection of the diffraction pattern along the c -axis ($hk0$ plane) at 297 and 50 K. The new peaks (marked by the yellow circle) appearing at 50 K become visible due to the distortion of the Co lattice.

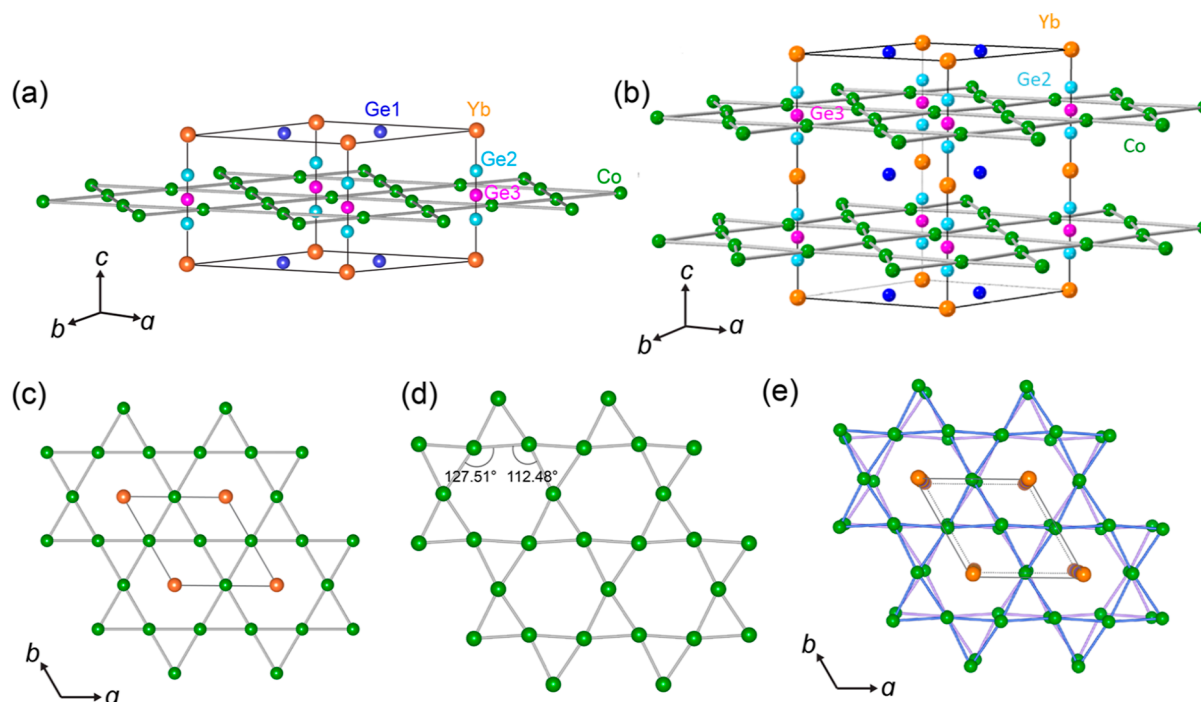


Figure 3. Crystal structures of $\text{Yb}_{0.5}\text{Co}_3\text{Ge}_3$ at 297 and 50 K. (a,c) Crystal structures at 297 K with the space group of $P6/mmm$ and projection down the c -direction of the Co Kagome network. (b) Crystal structure at 50 K with the space group of $P6_3/m$. (d) Distortion of one of the Co Kagome layers at 50 K with the modified bond angles of the hexagons labeled. (e) Two layers of the distorted Co Kagome layers unit cell showcasing their rotation relative to each other.

between the edges at neighboring vertices of the hexagons. This results in a deformation of the Kagome net, a twisting of the triangular units with respect to each other that breaks C_6 rotational and inversion symmetry, and results in the loss of all mirror planes parallel to the c -axis, subsequently reducing the space group to $P6_3/m$ from the original $P6/mmm$. Additionally, the geometric distortion of the Kagome subunit changes the

long-range ordering along the c -axis. At room temperature, the unit cell is defined with the shorter c -axis ($c \sim 3.91 \text{ \AA}$) and one Kagome subunit located at $z = 1/2$. At $T = 50 \text{ K}$, the long-range ordering along the c -axis requires a doubling ($c \sim 7.78 \text{ \AA}$) and the unit cell now contains two Kagome subunits located at $z = 1/4$ and $3/4$ (Figure 3b,e). These are related to each other by a sixfold screw axis along c , and an inversion

center is located between the Kagome nets, making the full structure centrosymmetric, even though a single distorted Kagome net breaks inversion symmetry (Figure 3d). The distorted Kagome net at low temperature is distinct from AV_3Sb_5 family,^{19,23} a similar distortion has been seen in the Kagome metals MgCo_6Ge_6 ,⁵⁰ LaRu_3Si_2 , and YRu_3Si_2 , and some of the phases are superconductors but show no sign of magnetic or CDW transitions above T_c .^{51–55} It is worth noting that although a magnetic transition at low temperature (18 K) was detected in the previous magnetic susceptibility study, no magnetic transition signal was observed between 50 and 300 K.⁴¹ This may be related to the distortion of the Co Kagome nets, which is inverted between the two layers and may influence magnetization; future experiments such a muon spin relaxation may help to explicitly understand the magnetism and its relation to the resistive transition in $\text{Yb}_{0.5}\text{Co}_3\text{Ge}_3$. The feature of a resistive transition arising from a CDW but without an associated magnetic signal has been observed in other materials.^{56,57}

In summary, we investigated the crystal structure and transport properties of the Kagome metal $\text{Yb}_{0.5}\text{Co}_3\text{Ge}_3$ and found a symmetry lowering structural phase transition around 95 K commensurate with a kink in the resistivity that is very similar to the CDW transition found in the AV_3Sb_5 and FeGe Kagome metals. Based on our measurements down to 2 K, we do not find intrinsic superconductivity; however, we do observe anisotropic negative magnetoresistance that correlates with previous study on a magnetic transition around 18 K. The 95 K transition distorts the Kagome net, keeping the triangular units consistent but rotating them slightly relative to each other, breaking C_6 rotation, out-of-plane mirror, and inversion symmetry. Since it is known that the structure of the Kagome lattice drives flat bands, Dirac bands, and magnetic frustration in Kagome metals, resulting in strong electron correlation, topological electrons, and complex magnetic states, which significantly influences the electronic properties, $\text{Yb}_{0.5}\text{Co}_3\text{Ge}_3$ and its distortion merit further theoretical and experimental investigation. Angle-resolved photoemission spectroscopy with associated band structure calculations can be performed to understand the band structure modifications associated with the phase transition and correlations in this material. Of particular importance is determining whether the structural transition is associated with the formation of the CDW order in analogy to other Kagome metals, ideally studied using scanning tunneling microscopy (STM). As recently discovered in the AV_3Sb_5 family, where multiple charge-ordered phases were found as a function of decreasing temperature via STM but were hidden in resistive transport measurements, there may be further orderings present in $\text{Yb}_{0.5}\text{Co}_3\text{Ge}_3$, which remain to be found. Additionally, the magnetism in $\text{Yb}_{0.5}\text{Co}_3\text{Ge}_3$ at low temperature may also induce spin orders, and muon spin relaxation measurements could elucidate this. Finally, since superconductivity has been observed in some other materials with distorted Kagome lattices and nematic superconductivity in the AV_3Sb_5 family, further studies based on chemical doping or high-pressure approaches at lower temperature can be used in $\text{Yb}_{0.5}\text{Co}_3\text{Ge}_3$ to probe this and its potential relation to the structural transition.

■ ASSOCIATED CONTENT

SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.chemmater.2c01309>.

CoSn and HfFe_6Ge_6 structure types and temperature dependence of resistivity of an additional sample with and without the field (PDF)

$\text{Yb}_{0.5}\text{Co}_3\text{Ge}_3$ at 50 K (CIF)

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Notes

The authors declare no competing financial interest.

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