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SPECIAL TOPIC — Structures and properties of materials under high pressure

High pressure growth of transition-metal monosilicide RhGe single crystals

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Transition-metal monosilicide RhGe has been reported to exhibit weak itinerant ferromagnetism, superconductivity, and topological properties. In this study, we report the high-pressure growth of high-quality RhGe single crystals up to millimeter size using a flux method. Transport measurements reveal metallic behavior in RhGe from 2 K to 300 K with Fermi liquid behavior at low temperatures. However, no superconductivity was observed with variations in the Ge composition. Magnetic characterizations indicate that RhGe exhibits paramagnetic behavior between 2 K and 300 K. The high-quality and large-size RhGe single crystals pave the way for further investigation of their topological properties using spectroscopic techniques.

Keywords: transition metals monosilicide, high pressure, single crystal growth

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1. Introduction

Transition-metal monosilicides *MX* (*M* = Cr, Mn, Fe, Co, Ru, Rh, and Os; *X* = Si and Ge), which crystallize in the FeSi-type structure (B20 structure, space group $P2_13$), featuring a cubic noncentrosymmetric lattice, have attracted significant attention recently due to their intriguing magnetic, topological, and transport properties.[1–6] For instance, MnSi exhibits an itinerant helical magnetic structure below T_C = 30 K, transitioning to a conical structure under a magnetic field and eventually evolving into ferromagnetism.[7–10] Additionally, a skyrmion phase has been observed in the conical phase near T_C.[11,12] This magnetic structure can be suppressed under pressure, with a quantum critical point observed at approximately 1.46 GPa.[8,9] Another notable compound, CoSi, demonstrates a nontrivial electronic structure topology.[13–15] The electronic structure with nonzero topological charge and Fermi arcs, connecting projections of band-touching nodes at Γ and R points on the surface Brillouin zone, has been observed in angle-resolved photoemission spectroscopy (ARPES) experiments.[16–18] Besides their fascinating magnetic and topological properties, transition metals monosilicides also exhibit excellent thermoelectric performance. For example, CoSi shows a Seebeck coefficient at room temperature of about −80 μV/K at stoichiometric composition.[19] A power factor of up to 5.5 mW/mK^2 has been observed in CoSi at 350–400 K, surpassing that of the well-known thermoelectric material Bi_2Te_3.[19]

Despite the exotic physical properties discovered in B20-structured materials, superconductivity remains rare. Tsvyashchenk *et al.*[20] reported that RhGe showed a superconducting state below 4.3 K and weak itinerant-electron ferromagnetism below 140 K. However, no further investigations have been conducted on this compound since then. The absence of inversion symmetry in the superconducting order parameter results in a mixture of spin-singlet and spin-triplet components,[21] making RhGe a possibly novel platform for studying p-wave superconductivity. Moreover, the potential coexistence of itinerant ferromagnetism and superconductivity in RhGe provides a unique opportunity to explore the intrinsic physical properties of unconventional superconductivity.[20] Recent ARPES experiments have observed band-touching

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nodes in the bulk electronic spectrum and Fermi arcs in CoSi.[16–18] However, the small spin–orbit coupling (SOC) in CoSi makes it challenging to resolve closely lying pairs of Fermi arcs experimentally, and the energy positions of these nodes and the peculiarities of the corresponding surface Fermi arcs must be determined through band structure calculations.[16–18] Given that the SOC of RhGe is significantly larger than that of CoSi, with node splitting nearly twice as large,[2] RhGe also offers an opportunity to study the evolution of band structures and Fermi arcs with the enhancement of SOC.

To address these issues, high-quality RhGe single crystals are essential. High-quality single crystals are indispensable for determining electronic structures using ARPES, and they often reveal intrinsic properties that cannot be observed in polycrystalline samples. In this work, we report the growth of RhGe single crystals under high pressure and high temperature, along with detailed characterization of their physical properties.

2. Experimental details

Single crystals of RhGe were grown using a high-pressure cubic anvil apparatus. The precursors of Rh (99.99%) and Ge (99.99%) with a mole ratio of 1:2 were thoroughly ground and mixed. Additional Ge was added as a flux due to its low melting point. The precursor was sealed into an hBN capsule (since Au and Pt capsules were avoided as they can react with Ge under high pressure and high temperature), then placed into the center of the high-pressure cells and compressed up to 5 GPa. The temperature was first rapidly increased to 1200 °C with a rate of 6 °C/min and maintained at that temperature for 4 hours, followed by a gradual cooling to 600 °C over 30 hours.

Finally, the sample was quenched to room temperature before releasing the pressure. The single-crystal quality and orientation were verified using single-crystal x-ray diffraction on a high-resolution system (SmartLab) with Cu K_α radiation ($\lambda = 1.5406$ Å), and the Laue pattern was collected in backscattering mode. Powder x-ray diffraction was performed at room temperature, using an x-ray diffractometer (D/MAX 2500V) with Cu K_α radiation at operating voltage and current of about 40 kV and 150 mA, respectively. The structure was refined using the Rietveld method via the general structure analysis system (GSAS) program.[22] The magnetic properties were measured using the MPMS3 of Quantum Design under a magnetic field of 1 T in zero field cooling and field cooling modes. Scanning electron microscopy (SEM) was carried out by JEOL, JSM-7000F. Chemical compositions of single crystals were determined using a scanning electron microscope with an energy dispersive x-ray analyzer. Transport measurement was performed using the four-probe method. Specific heat was measured using a physical property measurement system (PPMS).

3. Results and discussion

Given the low melting point of the Ge element, we chose Ge as the flux to grow RhGe single crystals. Initial attempts with a Rh:Ge ratio of 1:1 yielded single crystals up to $1.0\times0.5\times0.5$ mm^3 in size, but the quality was suboptimal. By adjusting the initial ratio to 1:2 to increase the content of flux, we successfully obtained high-quality RhGe single crystals. Figure 1(a) shows a photograph of single crystals of RhGe up to millimeter size. It can be seen that the crystals exhibit a dark color with a shiny surface (see Fig. 1(a)). X-ray powder diffraction can be well fitted using the $P2_13$ structure (see Fig. 1(b)). Energy-dispersive x-ray (EDX) analysis confirmed the composition of Rh and Ge to be approximately 1:1, which is in excellent agreement with the nominal composition (see Fig. 1(c)). Figure 1(d) shows a Laue back reflection pattern on a RhGe single crystal for a highly symmetrical plane of (100). Sharp diffraction spots were observed without visible spallation, and all the spots were well-indexed based on cubic symmetry, suggesting the high quality of RhGe single crystals.

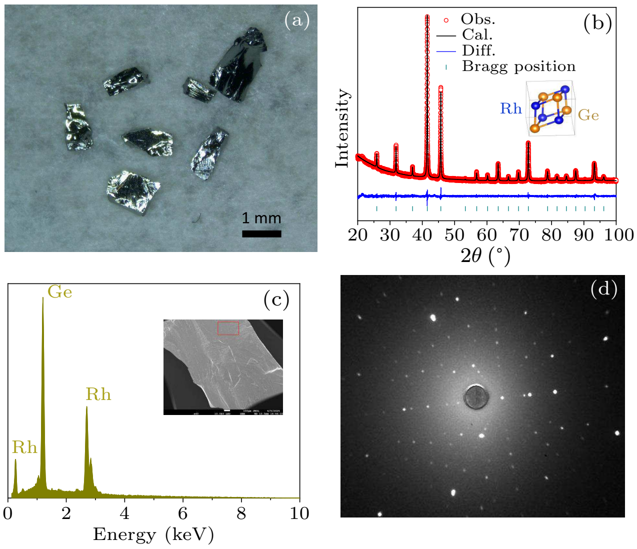

Fig. 1. (a) Photograph of RhGe single crystal. (b) Rietveld refinement of the x-ray diffraction pattern of RhGe. The inset shows the crystal structure of RhGe. (c) EDX spectrum for RhGe single crystal. The inset shows the SEM image of this sample. (d) Laue diffraction spots of the (100) plane for RhGe single crystal.

The resistivity of RhGe single crystal $\rho(T)$ exhibits metallic behavior between 25 K and 300 K, with a gradual upturn below 25 K, possibly indicating weak localization. No drop indicating the onset of superconductivity was observed. The low-temperature range of $\rho(T)$ curve between 25 K and 70 K follows the T^2 dependence, consistent with Fermi liquid behavior, rather than the T^3 behavior reported in previous studies. As no superconductivity was observed down to 2 K, we believe its absence of superconductivity may be intrinsic and not due to variations in Ge content as previously reported.

Next, we focus on the magnetism of RhGe single crystals. The temperature dependence of magnetic susceptibility, $\chi(T)$, at a magnetic field of 1 T indicates that the RhGe single crystal exhibits paramagnetism in the whole temperature range (see Fig. 3(a)), and the ferromagnetism with T_{C} $\sim$ 140 K[20] is absent. Moreover, field-dependent magnetization curve, $M(\mu_0 H)$, at 5 K shows a nearly linear behavior (see Fig. 3(b)), indicating no magnetic order at low temperatures, consistent with paramagnetic behavior shown in $\chi(T)$ curve.

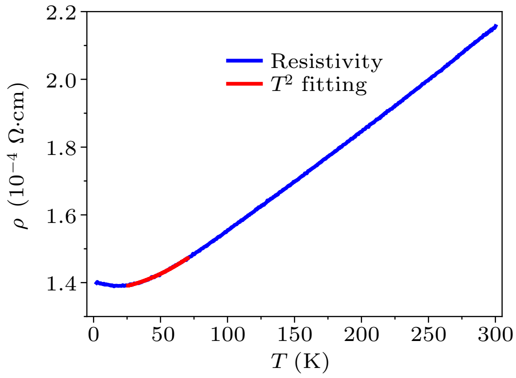

Fig. 2. Temperature dependence of resistivity for the RhGe single crystal. The red line shows the fit using the formula $\rho = \rho_0 + AT^2$.

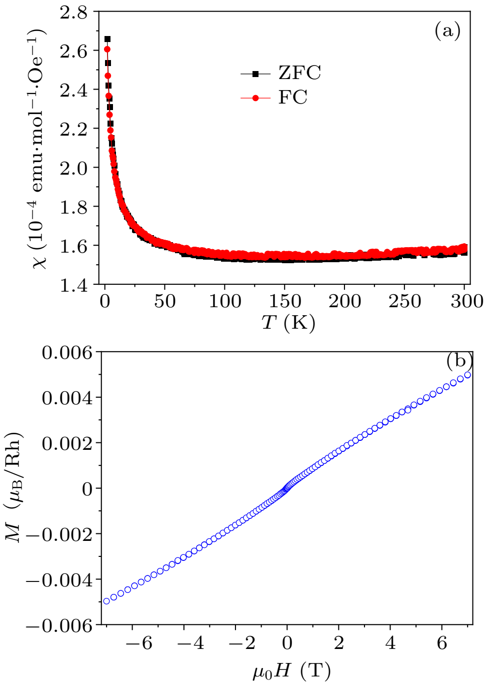

Fig. 3. (a) Temperature dependence of magnetic susceptibility $\chi(T)$ for the RhGe single crystal under a magnetic field of 1 T in zero-field-cooling and field-cooling modes. (b) Field-dependent magnetization $M(\mu_0 H)$ of the RhGe single crystal at 5 K.

Specific heat measurement of the RhGe single crystal is shown in Fig. 4. No significant jump either at 4.3 K or 140 K was observed. This result further confirms the absence of superconductivity and ferromagnetism in RhGe single crystals.

Since no superconductivity was observed in pristine RhGe, carrier doping or isovalent substitution may be possible ways to realize superconductivity in this system. Previous studies show that the orthorhombic phase IrGe exhibits superconductivity with $T_{\mathrm{c}} \sim 4.7$ K.[23] It would be interesting to study whether Ir doping in RhGe can induce superconductivity. Besides superconductivity, the larger SOC of Ir may also influence the surface states, potentially leading to novel topological properties. These studies will provide further opportunities to gain deeper insights into the intrinsic properties of B20-structured materials.

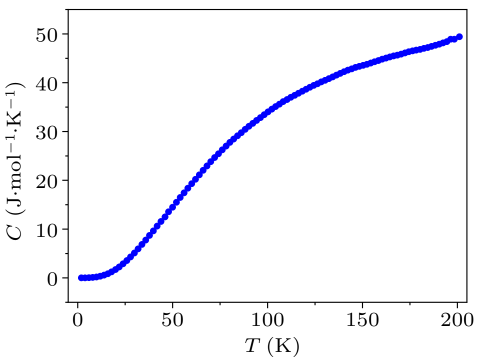

Fig. 4. Temperature dependence of specific heat for the RhGe single crystal.

4. Summary

We successfully grew high-quality RhGe single crystals up to millimeter size under high pressure and high temperature. Transport measurements reveal the metallic behavior and Fermi liquid behavior of RhGe single crystals at low temperature, while magnetic measurements indicate that RhGe exhibits paramagnetic behavior. Neither superconductivity nor ferromagnetism was observed down to 2 K. These high-quality RhGe single crystals provide a promising platform for further investigation of their topological properties via ARPES. Moreover, this study highlights the ongoing challenge of discovering superconductivity in B20-structured compounds.

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