

Superconductivity under pressure across the phase transition in a rhenium-based high-entropy alloy

Greeshma C. Jose , Kallol Chakrabarty , and Yogesh K. Vohra ^{*}

Department of Physics, *University of Alabama at Birmingham*, Birmingham, Alabama 35294, USA



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$\text{Re}_{0.6}(\text{HfZrNbTi})_{0.4}$ is a hexagonal high-entropy alloy superconductor that serves as a model system for exploring the interplay of disorder, crystal structure, and superconductivity. We investigate its superconducting properties under pressures up to 56.6 GPa using electrical transport measurements. The superconducting transition temperature, T_c , exhibits a modest increase at low pressures in the hexagonal phase, likely associated with pressure-induced changes in the electronic structure and electron-phonon coupling, and then saturates as it approaches the previously reported hexagonal-to-bcc structural transition. Despite the associated volume collapse, T_c evolves smoothly across the transition region, likely reflecting the broad coexistence of the hexagonal and bcc phases over the transition pressure interval. Upon decompression, T_c is systematically lower than during compression, consistent with hysteresis of the structural transition and metastable retention of the high-pressure phase. The upper critical field, $H_{c2}(0)$, exhibits a weak nonmonotonic pressure dependence and remains well below the Pauli paramagnetic limit, consistent with conventional spin-singlet pairing. These results demonstrate that superconductivity in $\text{Re}_{0.6}(\text{HfZrNbTi})_{0.4}$ remains remarkably robust against both lattice compression and structural transformation, consistent with the disorder-averaged superconducting state proposed for high-entropy alloys.

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I. INTRODUCTION

High-entropy alloys (HEAs) are a class of materials that have attracted significant attention due to their compositional complexity and the unusual properties that emerge from it. Their behavior is often interpreted in terms of four core effects: the high-entropy effect, lattice distortion, sluggish diffusion, and the cocktail effect [1–3]. These mechanisms collectively stabilize metastable solid-solution phases and impart exceptional mechanical strength, thermal stability, and chemical resilience.

The observation of superconductivity in HEAs is particularly notable, as it demonstrates that a coherent superconducting state can persist despite strong chemical disorder. A seminal study by Koželj *et al.* established bulk type-II superconductivity in Ta-Nb-Hf-Zr-Ti HEAs, consistent with a phonon-mediated BCS mechanism [4]. Subsequent studies have shown that superconductivity in HEAs is robust against disorder and external perturbations such as pressure [5,6]. T_c depends sensitively on valence electron concentration (VEC) and exhibits behavior intermediate between crystalline and amorphous systems [5,7,8], while the pairing mechanism is generally consistent with conventional electron-phonon interactions [2,9–13]. These characteristics, combined with their intrinsic mechanical robustness and compositional tunability,

make HEAs attractive for technological applications, particularly in environments requiring both structural stability and superconducting functionality [11,14–16]. In this context, the hexagonal HEA $\text{Re}_{0.6}(\text{HfZrNbTi})_{0.4}$ is of particular interest because hexagonal structures are relatively uncommon among transition-metal HEAs. Related compositions in the Nb-Re-Hf-Zr-Ti system exhibit bulk superconductivity with T_c values of approximately 4–5 K and conventional pairing behavior [12,17,18], demonstrating that superconductivity remains robust in this material family despite strong chemical disorder.

Pressure provides a powerful means to tune the structural and electronic properties of HEAs and probe the interplay between disorder and superconductivity [19]. In a previous study, Sereika *et al.* investigated the same sample batch used in the present work and showed that, at ambient conditions, $\text{Re}_{0.6}(\text{HfZrNbTi})_{0.4}$ crystallizes in a complex hexagonal phase (space group $P6_3/mmc$) with 12 atoms in the crystallographic unit cell [20]. They further reported a pressure-induced hexagonal-to-body-centered cubic (bcc) transformation near 44 GPa, accompanied by a significant volume collapse and metastable retention of the bcc phase upon decompression. The alloy was also found to exhibit exceptional mechanical performance, with a nanoindentation hardness of 18.5 GPa and a Young's modulus of 277.4 GPa, demonstrating its multifunctional character and making it a compelling system for investigation under extreme conditions. In particular, the superconducting response across this structural transition remains largely unexplored.

Motivated by these considerations, we study the superconducting properties of $\text{Re}_{0.6}(\text{HfZrNbTi})_{0.4}$ under high pressure using electrical transport measurements in a diamond anvil cell. The pressure dependence of T_c is examined over a wide pressure range, including across the previously reported

^{*}Contact author: ykvohra@uab.edu

structural transition, and $H_{c2}(0)$ is determined at selected pressures. Particular attention is given to the evolution of superconductivity in the vicinity of the pressure-induced hexagonal-to-bcc transformation and to the extent to which the superconducting state responds to the accompanying structural changes. We also track the evolution of superconductivity during both compression and decompression in order to assess possible effects associated with the metastable retention of the high-pressure phase. These measurements provide insight into the pressure-dependent superconducting behavior of this highly disordered system and help clarify the relationship between superconductivity, structural evolution, and chemical disorder under extreme compression.

II. EXPERIMENTAL METHODS

The $\text{Re}_{0.6}(\text{HfZrNbTi})_{0.4}$ sample used in this study is from the same batch as reported previously, and details of the synthesis can be found in Ref. [20]. Electrical resistance was measured using a standard four-probe configuration with the quantum design electrical transport option (ETO) installed on a PPMS DynaCool system. The ETO employs low-frequency ac excitation and phase-sensitive (lock-in) detection, enabling sensitive resistance measurements while minimizing dc offsets, instrumental drift, and broadband noise. High-pressure experiments were performed using a Be-Cu diamond anvil cell (DAC) with 300 μm culet diamonds. Resistance measurements were carried out using an ac current of 0.2 mA at a frequency of 18.3 Hz. A stainless-steel gasket, initially 250 μm thick, was preindented to 42 μm , and a 120 μm hole was drilled. The gasket was insulated with a cBN-epoxy mixture, and a 60 μm -diameter sample chamber was manually formed in the insulating layer. Thin Pt foils cut into wedge-shaped electrodes were used as electrodes, and the $\text{Re}_{0.6}(\text{HfZrNbTi})_{0.4}$ crystal was placed in the center of the chamber in contact with four probes. Experiments were carried out under nonhydrostatic conditions. Pressures were determined at room temperature by ruby fluorescence spectroscopy [21], measured both before and after the low-temperature runs to confirm stability; no significant variation was observed. To determine the upper critical field in the superconducting state, electrical resistance data were taken under applied magnetic fields up to 9 T at selected pressures in the PPMS.

III. RESULTS AND DISCUSSION

The superconducting properties of $\text{Re}_{0.6}(\text{HfZrNbTi})_{0.4}$ were investigated using a standard four-probe configuration under pressures ranging from 2.1 to 56.6 GPa. Electrical resistance measurements were carried out over the temperature range 1.8–300 K. Figures 1(a) and 1(b) show the normalized electrical resistance in the vicinity of the superconducting transition, where superconductivity is identified by the drop in resistance to zero. Well-defined and relatively sharp superconducting transitions, together with a clear zero-resistance state, are observed across the entire pressure range. At 2.1 GPa, the transition begins at an onset temperature of 4.4 ± 0.1 K and reaches zero resistance at 3.8 ± 0.1 K. The transition width remains relatively unchanged up to the highest applied

pressure of 56.6 GPa, where the onset temperature increases to 4.9 ± 0.1 K and zero resistance is achieved at 3.9 ± 0.1 K. The small negative resistance observed immediately below the superconducting transition is attributed to nonuniform current distribution during the formation of superconducting percolation paths. Similar behavior has been reported previously in superconducting transport measurements and is generally regarded as an experimental artifact arising from current redistribution rather than an intrinsic negative resistivity [23–25]. The anomaly remains confined to the vicinity of the superconducting transition and disappears once the sample reaches the zero-resistance state.

The temperature dependence of the normal-state electrical resistance at zero magnetic field is shown in Fig. 1(c). The normal-state resistance exhibits a weak positive temperature coefficient, with a room-temperature resistance of approximately 19.9 m Ω at 2.1 GPa, indicative of weak metallic behavior. The residual resistivity ratio (RRR), defined as $R(300 \text{ K})/R(8 \text{ K})$, is 1.53 at 2.1 GPa. This relatively small value suggests significant atomic-scale disorder in the high-entropy alloy. Such low RRR values are consistent with those reported in other HEA superconductors and disordered intermetallic compounds, reflecting strong scattering arising from chemical disorder and a reduced electronic mean-free path [4,5,17]. This weak metallic behavior persists across the entire pressure range studied in this work.

The T_c was determined using 90%, 50%, and 10% resistance criteria. Figure 2 summarizes the pressure dependence of T_c . The measured T_c values are slightly lower than those reported for a closely related composition at ambient pressure [17], consistent with the known sensitivity of superconductivity in high-entropy alloys to elemental composition [5]. For the 90% criterion, T_c increases from 4.23 K at 2.1 GPa to a maximum of 4.80 K at 36.4 GPa. At higher pressures, T_c remains nearly constant with a slight tendency toward saturation, reaching 4.64 K at the highest measured pressure of 56.6 GPa. Although minor variations are present, the 50% and 10% criteria exhibit the same overall pressure evolution, confirming that the observed trend is independent of the choice of T_c criterion. Overall, the results reveal only a weak variation of T_c with pressure, characterized by a modest increase at low pressures followed by saturation above ~ 35 GPa. The onset of this saturation occurs somewhat below the ~ 44 GPa structural transition reported from high-pressure x-ray diffraction measurements [20], which may reflect differences in pressure determination, including pressure-marker positioning and pressure gradients. Remarkably, despite compression to 56.6 GPa and the previously reported pressure-induced hexagonal-to-bcc transformation accompanied by a $\sim 6\%$ volume collapse, T_c changes by only $\sim 13\%$ over the entire pressure range.

The evolution of T_c under pressure can be understood in the context of previous studies of high-entropy-alloy superconductors, which suggest that superconductivity is governed primarily by the electronic structure and electron-phonon coupling, while the role of composition is mediated through its influence on these factors [4,5]. Compression modifies interatomic distances, orbital overlap, and lattice dynamics, leading to the gradual increase in T_c observed within the low-pressure hexagonal phase. Consistent with the structural

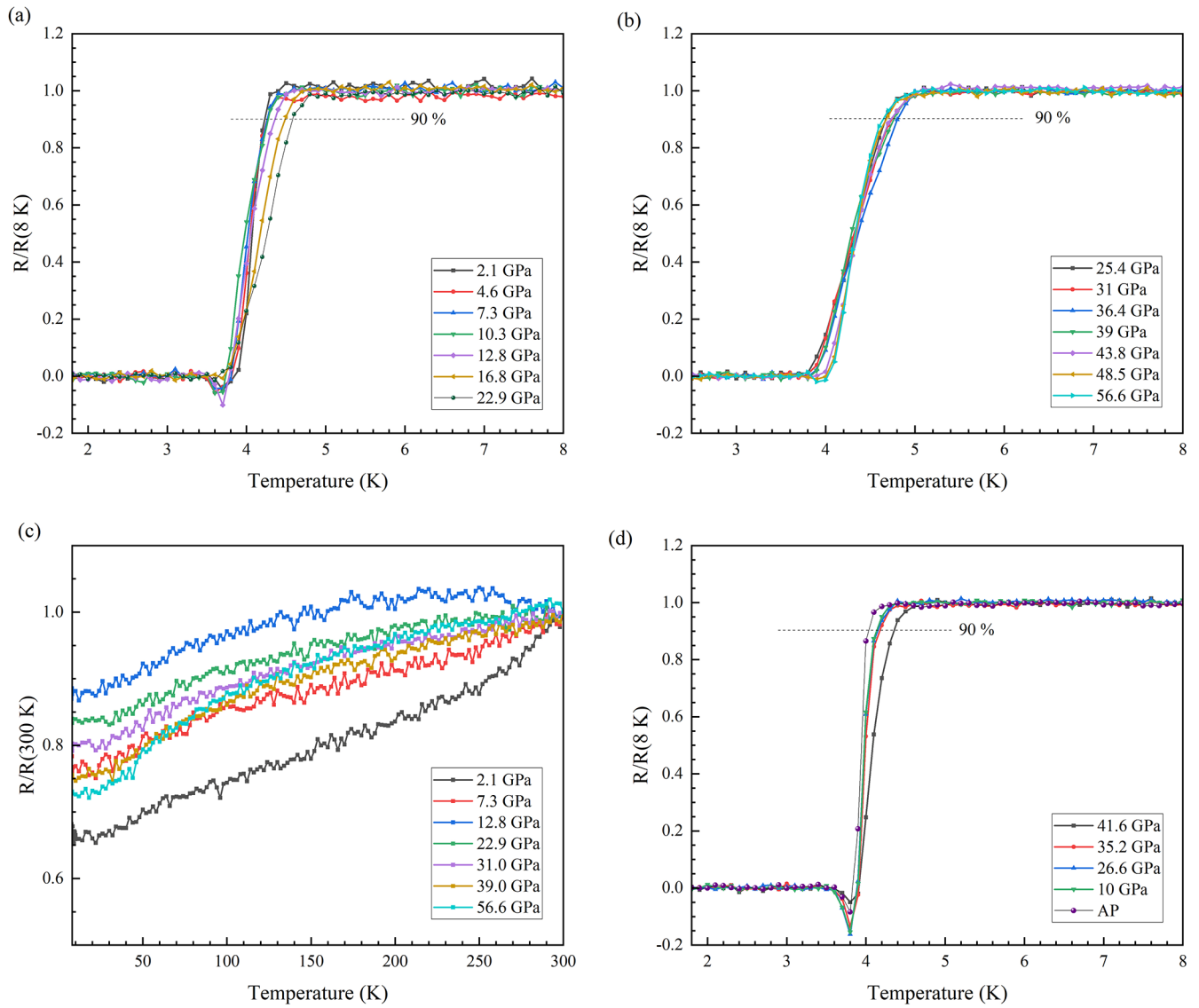


FIG. 1. Temperature-dependent electrical resistance of $\text{Re}_{0.6}(\text{HfZrNbTi})_{0.4}$ under pressures up to 56.6 GPa. (a), (b) Resistance as a function of temperature during compression. (c) Representative resistance versus temperature curves over the 300–10 K range at selected pressures. (d) Temperature-dependent resistance during decompression.

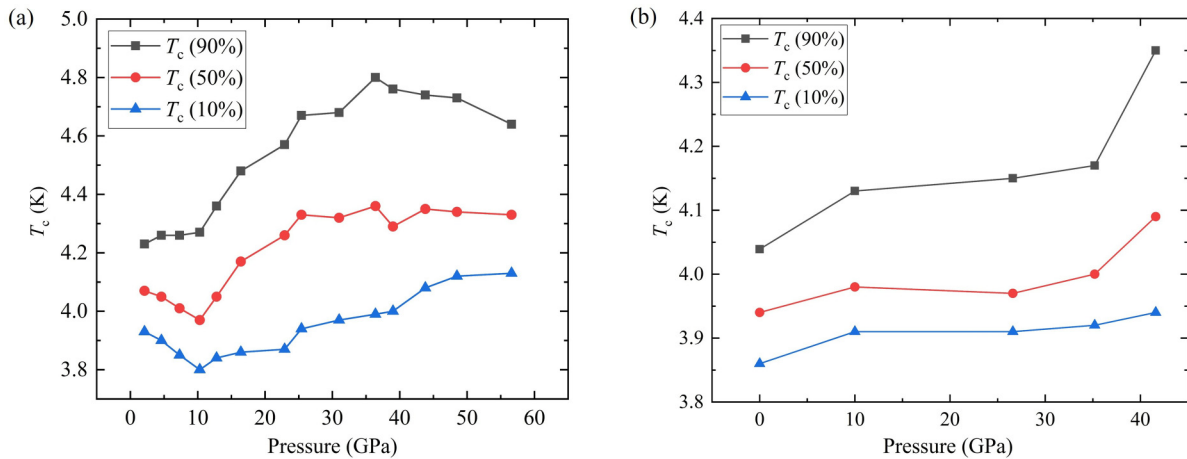


FIG. 2. Superconducting transition temperature, T_c , as a function of pressure. (a) Compression data showing T_c determined using the 90%, 50%, and 10% resistance criteria. (b) Decompression data showing T_c determined using the same 90%, 50%, and 10% resistance criteria.

results reported previously, the hexagonal lattice parameters decrease continuously with pressure, while the axial ratio (c/a) exhibits only a small variation [20]. The modest enhancement of T_c from 4.23 K at 2.1 GPa to 4.80 K at 36.4 GPa may arise from the corresponding pressure-induced changes in the electronic structure and electron-phonon coupling. In addition, the increase in T_c approaching the hexagonal-to-bcc transition may reflect enhanced electron-phonon coupling associated with the proximity to the structural instability, where phonon softening could enhance the superconducting pairing interaction.

The absence of an abrupt anomaly in T_c near the structural transition, despite the previously reported $\sim 6\%$ volume collapse, suggests that superconductivity is only weakly affected by the underlying lattice transformation. Previous high-pressure x-ray diffraction measurements showed that the hexagonal-to-bcc transition proceeds through a diffusionless martensiticlike mechanism and occurs over a finite pressure range in which both phases coexist [20]. In such a broad transition region, electrical transport measurements reflect an average response of coexisting phases, which may smooth any intrinsic changes associated with the structural transformation. Furthermore, both the hexagonal and bcc phases exhibit metallic transport behavior, and the relatively smooth evolution of T_c suggests that the electronic conditions supporting superconductivity remain broadly similar on both sides of the structural boundary. A large volume collapse does not necessarily imply a corresponding change in superconducting properties if the electronic states near the Fermi level and the average electron-phonon coupling strength evolve only moderately.

Beyond the effects of phase coexistence near the structural transition, the weak pressure dependence of T_c over the entire pressure range suggests that superconductivity in $\text{Re}_{0.6}(\text{HfZrNbTi})_{0.4}$ is remarkably robust against both lattice compression and structural evolution. Importantly, previous structural studies found no evidence of elemental segregation or long-range compositional rearrangement during the transformation, indicating that the chemically disordered solid-solution character of the alloy is largely retained. In conventional superconductors, pressure often produces changes in the density of states at the Fermi level, phonon frequencies, and electron-phonon coupling strength, which can lead to significant variations in T_c . In contrast, the strong chemical disorder and configurational complexity of high-entropy alloys generate a broad distribution of local atomic environments, causing superconductivity to emerge from an average over many local configurations rather than from a specific atomic arrangement [6,26–28]. The low residual resistivity ratio observed in the present alloy is consistent with significant atomic-scale disorder. Theoretical studies have suggested that superconductivity in HEAs behaves as a collective disorder-averaged state [29], thereby reducing the sensitivity of the superconducting pairing interaction to pressure-induced changes in the electronic structure and lattice dynamics. This interpretation is consistent with the present results, where T_c exhibits only modest variation with pressure despite compression to 56.6 GPa and a pressure-induced hexagonal-to-bcc transformation accompanied by a $\sim 6\%$ volume collapse. Although comparable electronic-structure calculations are not

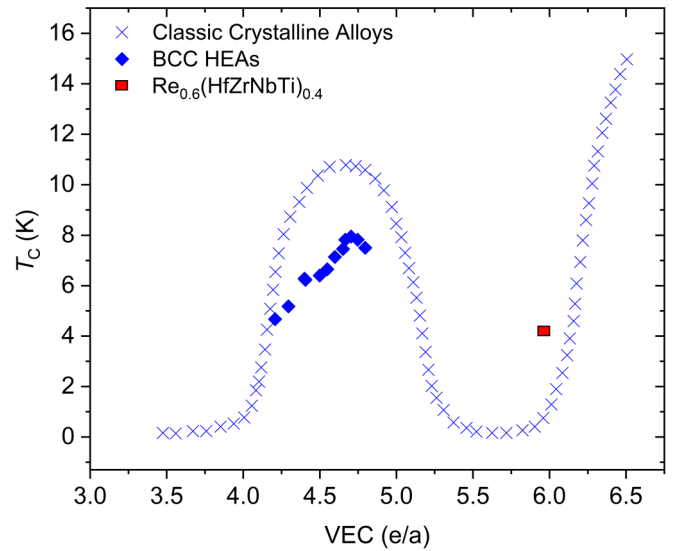


FIG. 3. Superconducting transition temperature, T_c , as a function of VEC. Blue crosses represent crystalline alloys, blue diamonds denote bcc HEA superconductors from the literature [2,5,22], and the red square marks bcc $\text{Re}_{0.6}(\text{HfZrNbTi})_{0.4}$.

currently available for $\text{Re}_{0.6}(\text{HfZrNbTi})_{0.4}$, the observed behavior is consistent with a superconducting state that remains largely insensitive to substantial lattice compression and structural evolution.

In the decompression cycle, T_c is systematically lower than the corresponding compression values at the same pressures. This behavior may be associated with the hysteresis of the hexagonal-to-bcc structural transition and the metastable retention of the high-pressure bcc phase reported previously [20]. Because the structural transition follows different paths during compression and decompression, the sample experiences a distinct structural history upon pressure release. In particular, decompression proceeds from the retained high-pressure bcc state, which persists to lower pressures, providing a plausible explanation for the offset between the compression and decompression T_c curves. Despite the reported retention of the bcc structure during decompression and upon recovery to ambient pressure, T_c does not retain its maximum high-pressure value and instead returns to approximately 4.04 K at ambient pressure. This observation suggests that the superconducting properties are influenced not only by the crystal structure but also by pressure-dependent changes in the electronic structure and lattice dynamics.

To place the superconducting properties of $\text{Re}_{0.6}(\text{HfZrNbTi})_{0.4}$ in the broader context of alloy superconductors, Fig. 3 compares its T_c with those of crystalline alloys and high-entropy alloys as a function of VEC. Classical crystalline alloys exhibit Matthias-type behavior, with a maximum in T_c near $\text{VEC} \sim 4.6\text{--}4.8$ and a reduction at higher electron concentrations [2]. bcc high-entropy alloys show qualitatively similar trends, although with generally lower T_c values due to the combined effects of chemical disorder and enhanced scattering. With $\text{VEC} \sim 5.9$ [20], $\text{Re}_{0.6}(\text{HfZrNbTi})_{0.4}$ lies in this higher-VEC regime and correspondingly exhibits a relatively modest T_c compared with lower-VEC superconducting alloys. The

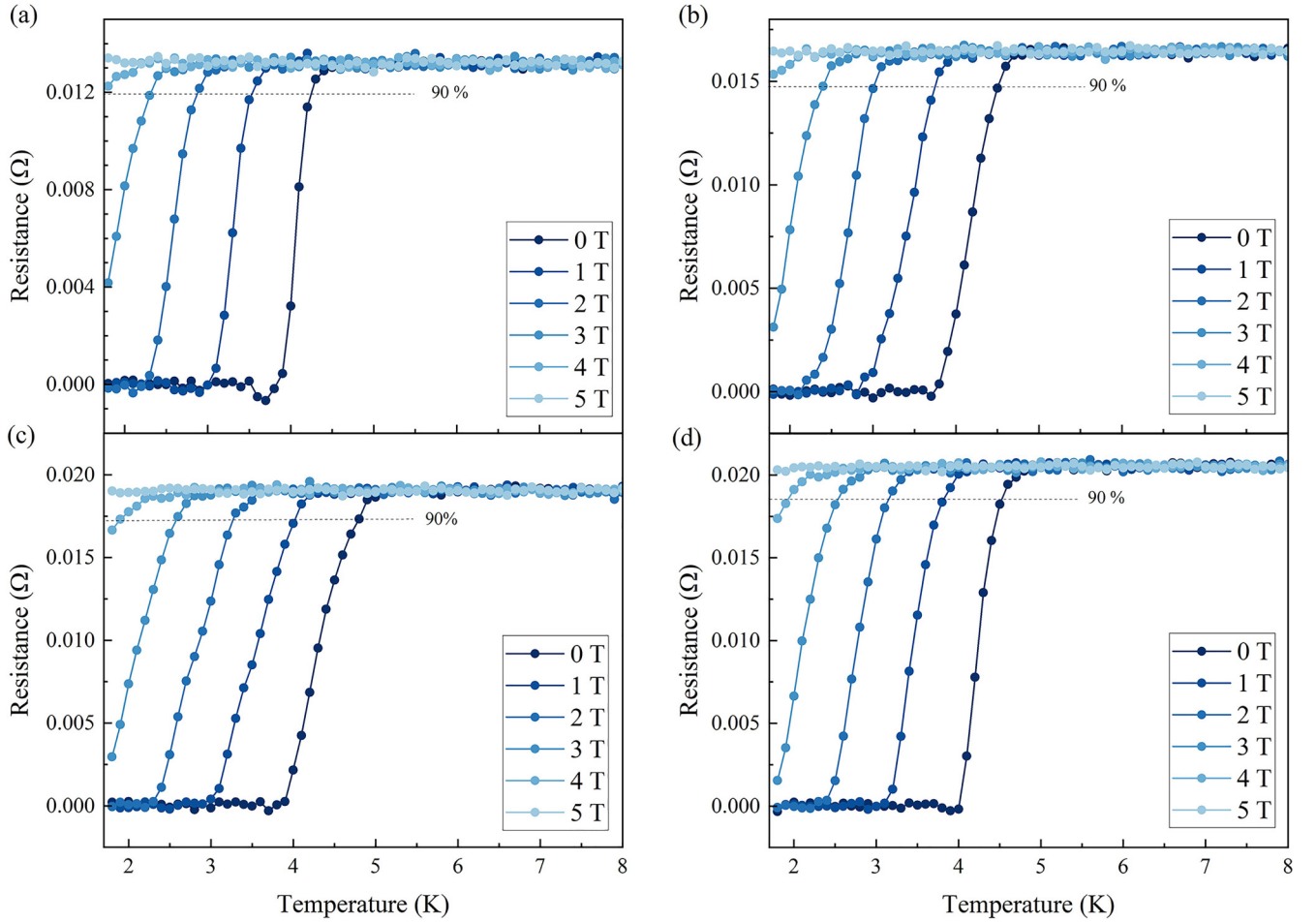


FIG. 4. Temperature-dependent resistance under applied magnetic fields at (a) 2.1 GPa, (b) 16.8 GPa, (c) 39.0 GPa, and (d) 56.6 GPa.

recovered ambient-pressure bcc phase of $\text{Re}_{0.6}(\text{HfZrNbTi})_{0.4}$ also falls within the higher-VEC region of the diagram consistent with the overall trend of modest superconducting transition temperatures observed among high-VEC HEA superconductors.

Figure 4 presents the temperature dependence of the normalized resistance under perpendicular magnetic fields up to 9 T, measured at 2.1, 16.8, 39.0, and 56.6 GPa, respectively, in the temperature range of 2–8 K. The normal-state resistance remains unaffected by the applied field, while the superconducting transition systematically shifts to lower temperatures with increasing magnetic field strength, consistent with superconducting behavior.

From Fig. 4, the T_c at different magnetic fields was extracted to construct the field-temperature phase diagram shown in Fig. 5. The orbital-limited upper critical field was estimated using the Werthamer-Helfand-Hohenberg (WHH) model:

$$H_{c2}^{\text{WHH}}(0) = -0.693 \left(\frac{dH_{c2}}{dT} \right)_{T=T_c} T_c. \quad (1)$$

The slopes of the $T_c(H)$ curves near T_c were used to estimate $H_{c2}^{\text{WHH}}(0)$ at each pressure. To describe the temperature dependence of the upper critical field over the full temperature

range, the Ginzburg-Landau (GL) expression was employed:

$$H_{c2}(T) = H_{c2}(0) \frac{1 - (T/T_c)^2}{1 + (T/T_c)^2}. \quad (2)$$

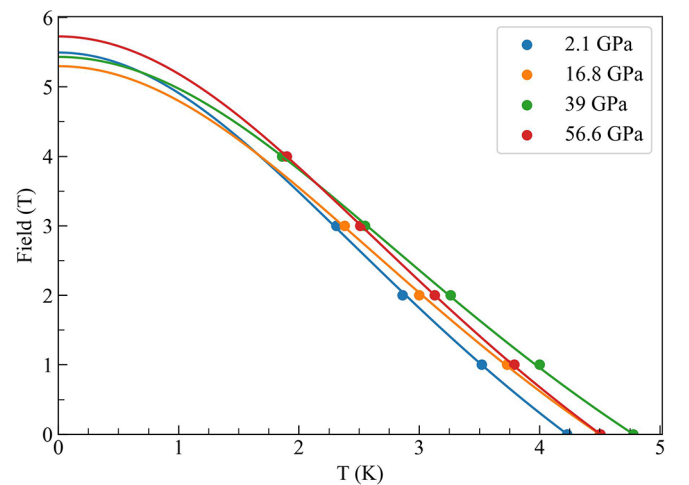


FIG. 5. Upper critical field, H_{c2} , as a function of temperature at selected pressures of 2.1, 16.8, 39.0, and 56.6 GPa. The H_{c2} values were extracted from electrical resistance measurements using the 90% resistance criterion. Solid lines represent fits to Eq. (2).

TABLE I. Upper critical field values of $\text{Re}_{0.6}(\text{HfZrNbTi})_{0.4}$ obtained from GL fitting, WHH analysis, and the Pauli paramagnetic limit $\mu_0 H_{c2}^{\text{Pauli}} = 1.86 T_c$ at different pressures.

P (GPa)	$H_{c2}^{\text{GL}}(0)$ (T)	$H_{c2}^{\text{WHH}}(0)$ (T)	H_{c2}^{Pauli} (T)
2.1	5.49	4.28	7.86
16.8	5.30	4.16	8.37
39.0	5.43	4.36	8.89
56.6	5.72	4.54	8.37

A summary of these parameters is presented in Table I. The $H_{c2}(0)$ values obtained from Ginzburg-Landau (GL) fitting are systematically higher than those estimated using the Werthamer-Helfand-Hohenberg (WHH) model. This difference arises because the GL expression extrapolates the full temperature dependence of $H_{c2}(T)$, whereas the WHH model is based on the initial slope near T_c and assumes orbital-limited behavior in the dirty limit. Both GL and WHH estimates of the upper critical field remain well below the Pauli paramagnetic limit, $\mu_0 H_{c2}^{\text{Pauli}} = 1.86 T_c$ [30], at all applied pressures. This suggests that superconductivity in $\text{Re}_{0.6}(\text{HfZrNbTi})_{0.4}$ is not primarily limited by Pauli pair breaking and is consistent with conventional spin-singlet pairing, as commonly observed in phonon-mediated superconductors in the weak to intermediate coupling regime. The relatively high values of H_{c2} and its temperature dependence further support the classification of this system as a type-II superconductor. However, a definitive determination of the pairing mechanism and coupling strength would require complementary thermodynamic or spectroscopic measurements.

The pressure dependence of the upper critical field differs somewhat from that of T_c . While T_c exhibits only a weak pressure dependence, with a modest increase followed by saturation at higher pressures, the estimated $H_{c2}(0)$ values exhibit a weak nonmonotonic variation with pressure and do not scale directly with T_c . This suggests that the parameters governing H_{c2} —including the coherence length, impurity scattering, and orbital limiting effects—evolve differently under pressure than those controlling T_c [31,32]. Since H_{c2} depends not only on the pairing strength but also on normal-state electronic properties and pair-breaking mechanisms, its pressure evolution need not mirror that of T_c .

IV. CONCLUSIONS

In summary, we have investigated the superconducting properties of $\text{Re}_{0.6}(\text{HfZrNbTi})_{0.4}$ under pressures up to 56.6 GPa using electrical transport measurements. The superconducting transition temperature, T_c , exhibits only a modest pressure dependence, increasing from 4.23 K at 2.1 GPa to a maximum of 4.80 K at 36.4 GPa before saturating at higher pressures. The small enhancement of T_c in the low-pressure hexagonal phase may be associated with pressure-induced modifications of the electronic structure and electron-phonon coupling. Despite the previously reported pressure-induced hexagonal-to-bcc transformation accompanied by a $\sim 6\%$ volume collapse, T_c changes by only $\sim 13\%$, indicating that superconductivity is only weakly affected by the underlying structural evolution.

The smooth evolution of T_c across the transition region may be related to the broad pressure interval over which the hexagonal and bcc phases coexist, causing transport measurements to probe an averaged response of the two phases. The weak overall variation of T_c is also consistent with the disorder-averaged nature of superconductivity proposed for high-entropy alloys, highlighting the remarkable robustness of the superconducting state against both lattice compression and structural transformation. During decompression, T_c is systematically lower than during compression, consistent with the hysteresis of the structural transition and the metastable retention of the high-pressure bcc phase. The upper critical field, $H_{c2}(0)$, exhibits a weak nonmonotonic variation with pressure distinct from that of T_c and remains well below the Pauli paramagnetic limit, consistent with conventional spin-singlet superconductivity. These results provide insight into the interplay among disorder, crystal structure, and superconductivity in high-entropy alloy superconductors.

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DATA AVAILABILITY

The data that support the findings of this article are available on request.

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