Supporting Information for

High-Entropy Alloy Superconductors on an α-Mn lattice

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SEM images of $(HfTaWPt)_{0.40}[Re]_{0.60}$

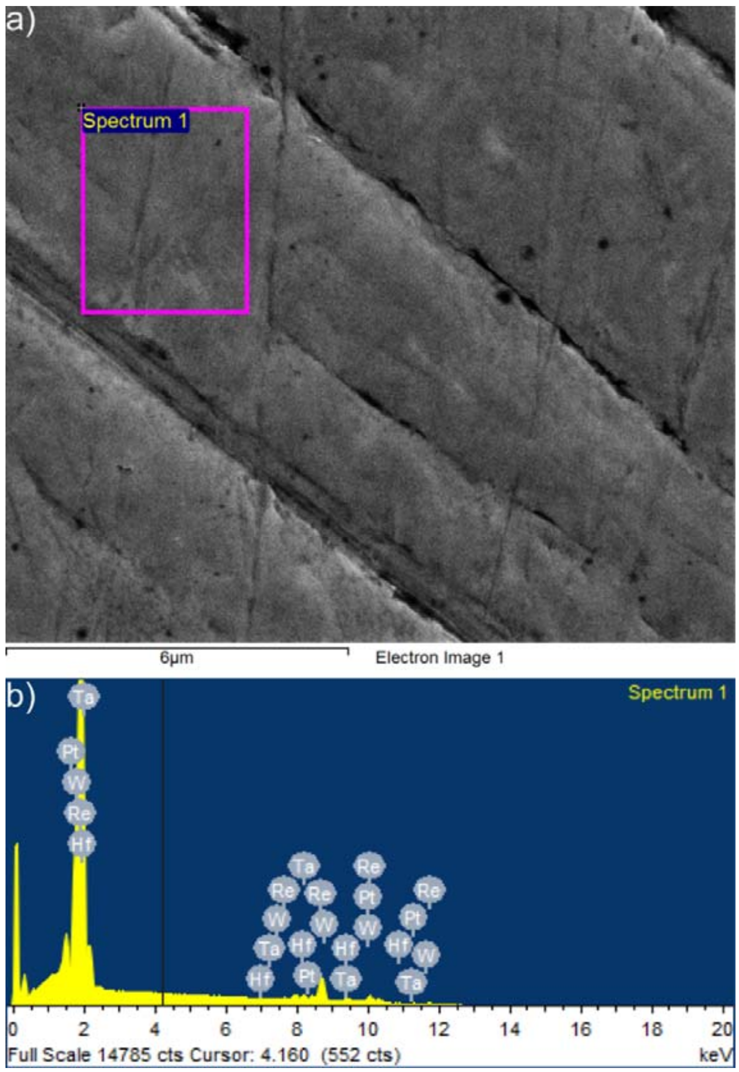

Figure S1. a) SEM image of the smooth surface of an as-cast, sanded $(HfTaWPt)_{0.40}[Re]_{0.60}$ sample; b) EDX spectrum of an area, emphasized in a), revealing all five elements present with stoichiometry in agreement with the initial composition.

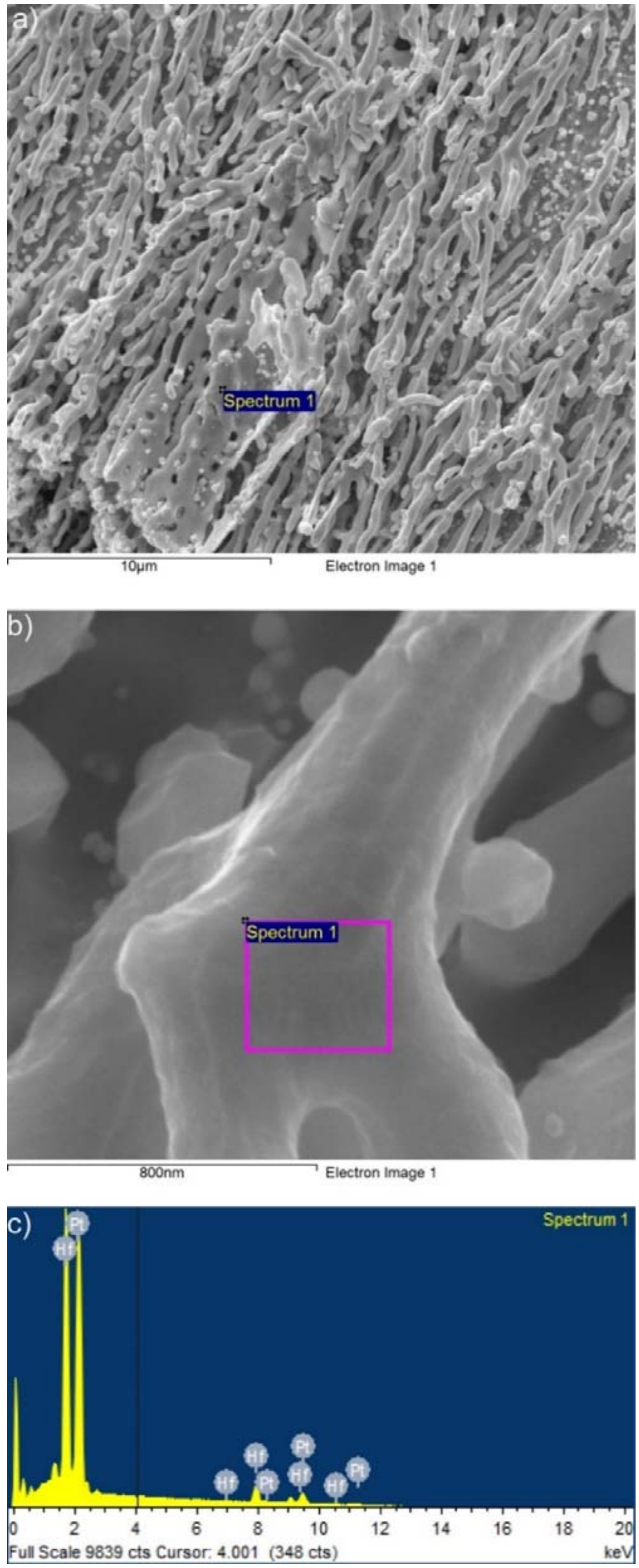

Figure S2. a) SEM image of the dendritic surface of a decomposed $(HfTaWPt)_{0.40}[Re]_{0.60}$ sample; b) SEM image with increased magnification of a sub-microscale dendrite; c) EDX spectrum of a selected area, emphasized in b), revealing a Hf/Pt-rich composition.

Heat capacity parameterization

The temperature dependent specific heat capacity can be expressed by the equation

$$\frac{C(T)}{T} = \gamma + \beta T^2 \quad (1),$$

where $\gamma \cdot T$ describes the electronic and $\beta \cdot T^3$ the phonon contribution to the heat capacity. The Sommerfeld parameter γ, and β, can be determined by the linear fit of the C/T vs. T^2 of the normal-state data collected with an applied magnetic field of 9 T (Figure S3a–c). The Debye temperature Θ_D was calculated using β according to the equation

$$\Theta_D = \left(\frac{12\pi^4}{5\beta} nR\right)^{\frac{1}{3}} \quad (2),$$

where the number of atoms per formula unit is $n = 1$ and the gas constant $R = 8.314$ $Jmol^{-1}K^{-1}$. The Debye temperatures for $(ZrNb)_{0.10}[MoReRu]_{0.90}$, $(HfTaWIr)_{0.40}[Re]_{0.60}$, and $(HfTaWPt)_{0.40}[Re]_{0.60}$ amount to 339 K, 317 K, and 252 K, respectively. The heat capacity parameterization for all three HEAs are summarized in Table S1.

Resistivity and upper critical fields

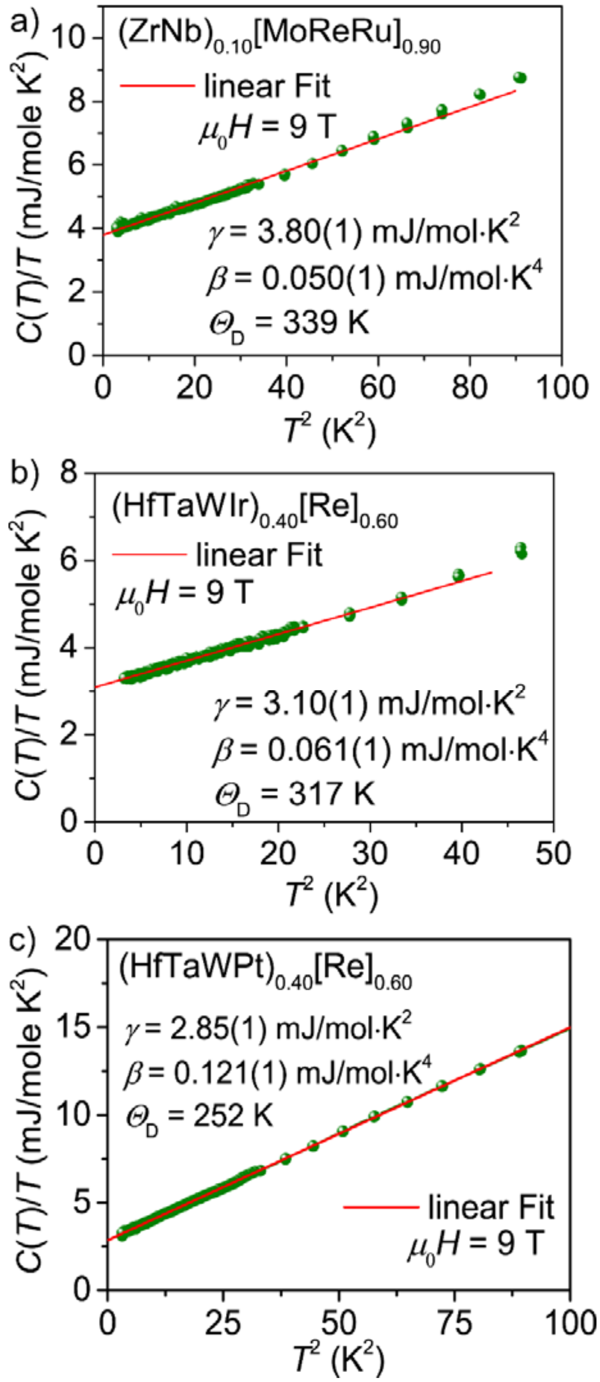

Figure S3. C/T vs. T^2 for a) $(ZrNb)_{0.10}[MoReRu]_{0.90}$, b) $(HfTaWIr)_{0.40}[Re]_{0.60}$, and c) $(HfTaWPt)_{0.40}[Re]_{0.60}$ measured in $\mu_0 H = 9$ T (normal state) fitted to $C/T = \gamma + \beta\, T^2$ (red line).

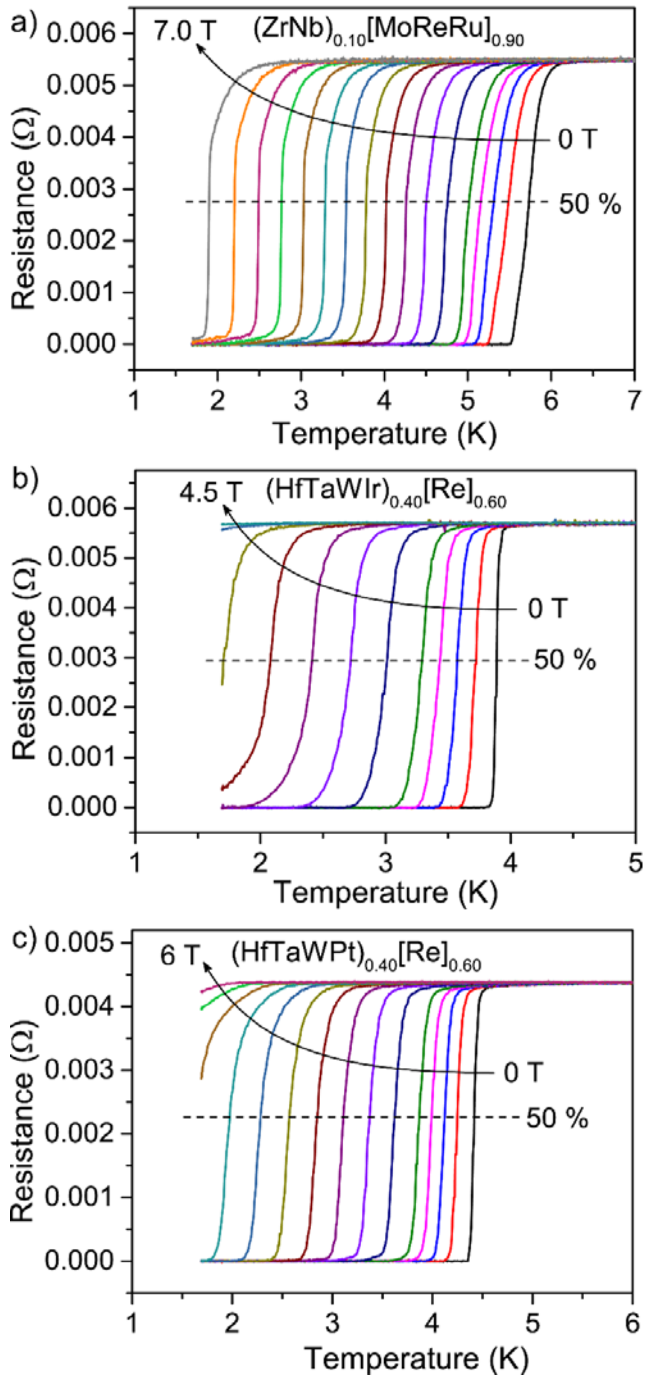

Figure S4. Magnetic field dependence of the superconducting transition for a) $(ZrNb)_{0.10}[MoReRu]_{0.90}$, b) $(HfTaWIr)_{0.40}[Re]_{0.60}$, and c) $(HfTaWPt)_{0.40}[Re]_{0.60}$ for 0 T $\leq \mu_0 H \leq$ 7.5 T, 0 T $\leq \mu_0 H \leq$ 4.5 T, and 0 T $\leq \mu_0 H \leq$ 6 T respectively; 50%-criterion depicted as black dashed lines.

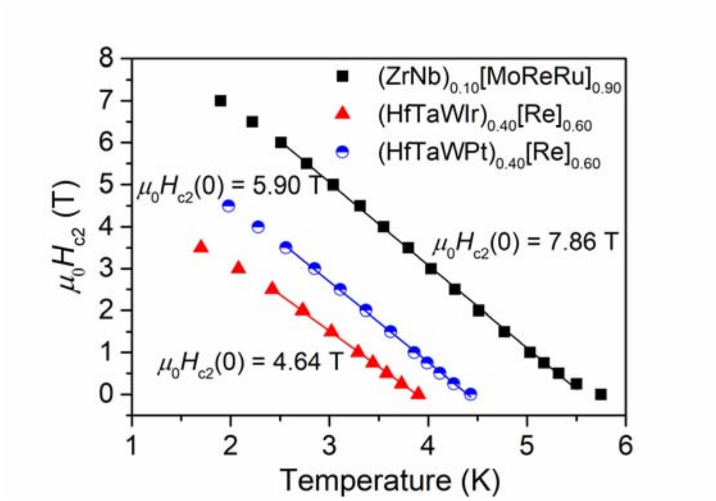

Figure S5. Temperature dependent upper critical fields $\mu_0H_{c2}(T)$ of $(ZrNb)_{0.10}[MoReRu]_{0.90}$, $(HfTaWIr)_{0.40}[Re]_{0.60}$, and $(HfTaWPt)_{0.40}[Re]_{0.60}$ HEAs. The open circles are the 50%-values obtained from $\rho(T)$ plots at different applied fields; the lines show the linear fits used to determine $\mu_0H_{c2}(0)$ according to the WHH approximation.

The linear regression gave very good fits to all experimental data (R^2: 0.9997, 0.9991, and 0.9994 for $(ZrNb)_{0.10}[MoReRu]_{0.90}$, $(HfTaWIr)_{0.40}[Re]_{0.60}$, and $(HfTaWPt)_{0.40}[Re]_{0.60}$, respectively). The new α-Mn-type HEA $(ZrNb)_{0.10}[MoReRu]_{0.90}$ exhibits the highest $T_c \approx 5.3$ K as well as the highest zero temperature upper critical field $\mu_0H_{c2}(0) = 7.86$ T among the materials studied here.

Table S1. Overview of the superconducting parameters obtained from susceptibility, specific heat and resistivity measurements for the $(ZrNb)_{0.10}[MoReRu]_{0.90}$, $(HfTaWIr)_{0.40}[Re]_{0.60}$, and $(HfTaWPt)_{0.40}[Re]_{0.60}$ HEA superconductors.

Parameter	$(ZrNb)_{0.10}[MoReRu]_{0.90}$	$(HfTaWIr)_{0.40}[Re]_{0.60}$	$(HfTaWPt)_{0.40}[Re]_{0.60}$
$T_{c,\ magnetization}$ (K)	5.3	4.0	4.4
$T_{c,\ specific\ heat}$ (K)	5.3	4.0	4.4
$T_{c,\ resistivity}$ (K)	5.74	3.90	4.43
γ (mJ·mol^{-1}·K^{-2})	3.80(1)	3.10(1)	2.85(1)
β (mJ·mol^{-1}·K^{-4})	0.050(1)	0.061(1)	0.121(1)
Θ_D (K)	339	317	252
$\Delta C/\gamma T_c$	1.53	1.46	1.46
ρ_{300K} (μΩ·cm)	105	536	591
RRR	1.02	1.01	1.01
$\mu_0 H_{c2}(0)$ (T)	7.86	4.64	5.90