Superconductivity in single crystals of $Lu_3T_4Ge_{13-x}$ (T = Co, Rh, Os) and $Y_3T_4Ge_{13-x}$ (T = Ir, Rh, Os) Supporting Information (SI)

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I. ADP RATIO CALCULATIONS

The equation used to determine each materials' ADP ratio is given by: ADP ratio = $U_{eq}(2a)/U_{eq}(\mathrm{T})$ where $U_{eq}(2a)$ is the displacement parameter for the atom located at Wyckoff site $2a$, and $U_{eq}(\mathrm{T})$ is the displacement parameter for the transition metal element located at Wyckoff site $8e$. In the case of displacement parameters in literature being reported in B_{iso} or B_{eq} rather than U_{eq}, the following equation was used to relate $B_{iso/eq}$ to U_{eq}: $B_{iso/eq} = 8\pi^2(U_{eq})$.

II. PHYSICAL PROPERTIES MEASUREMENTS

The specific heat $\mathrm{C}_p(T)$ for $\mathrm{Lu_3}T_4\mathrm{Ge}_{13-x}$ (T = Co, Rh, Os) and $\mathrm{Y_3Os_4Ge}_{13-x}$ measured in various applied magnetic fields (Fig. S1) confirms the superconducting ground state these compounds. As the magnetic field increases, the transition is suppressed below 0.4 K for H between 1.4 T and 3.4 T. Linear fits (dashed lines in Fig. S1e) of the normal state specific heat, C_p/T *vs.* T^2, for $\mathrm{T} > \mathrm{T}_c$ yield very similar electronic and phonon specific heat coefficients $\gamma_n \approx 5$ - 8 $\mathrm{mJ/molK^2}$ for the four compounds (Table S3). After subtracting the phonon contribution, the resulting $H = 0$ electronic specific heat scaled by temperature C_e/T is plotted in Fig. 4. Equal entropy constructs (dashed lines) yield an estimate for the electronic specific heat jump at T_c, $\Delta C_e/\gamma T_c$ close to 1.43 for the two Os compounds (Fig. 4c-d), and smaller (close to 1) for the other two compounds (Fig. 4a-b).

When considering a residual electronic specific heat $\mathrm{C}_{e,0} = \gamma_0\mathrm{T}$ (as introduced in the main text),[1] the superconducting gap $2\Delta(0)/k_BT_c$ can be estimated from the fit $C_e = \gamma_0 T + T_c A e^{-\Delta(0)/k_BT}$. These fits with $T < 0.4\ T_c$, with an example shown as dashed lines in Fig. 4, yield $2\Delta(0)/k_BT_c$ values between 2.95 and 3.12 (Table S3). The $2\Delta(0)/k_BT_c$ values, smaller than the predicted BCS value of 3.53, suggest that these are weakly coupled superconductors. The above superconducting gap energy equation is valid for the fits much lower than T_c, $T \leq 0.4\ T_c$.[2] With the limited temperature range available below $0.4\mathrm{T}_c$ in these compounds, it is very difficult to discern, from the C_e fits alone, whether impurity or multi-band superconducting effects are at play. Such a reduced energy gap ($2\Delta(0)/k_BT_c = 1.7$) was also reported for single crystals of $\mathrm{Y_3Ru_4Ge_{13}}$,[3] an isostructural 3-4-13 compound with a similarly small specific heat jump at T_c. As the resistivity data shows (Fig. 5 main text), one more similarity with the $\mathrm{Y_3Ru_4Ge_{13}}$ is the semiconductor-*like* resistivity in the normal state, a common trait of the 3-4-13 germanide superconductors, but distinct from the good metal behavior in all other 3-4-13 superconductors.[4,5]

The temperature-dependent Hall coefficient R_H measured for $H = 9$ T is shown in Fig. S2 for $\mathrm{Lu_3}T_4\mathrm{Ge}_{13-x}$ (T = Co, Rh, Os) and $\mathrm{Y_3Os_4Ge}_{13-x}$. $\mathrm{Lu_3Rh_4Ge_{13}}$ (Fig. S2a) and $\mathrm{Y_3Os_4Ge}_{12.65}$ (Fig. S2d) show both types of charge carriers, as demonstrated by the sign change of R_H. In $\mathrm{Lu_3Co_4Ge_{13}}$ (Fig. S2b) and $\mathrm{Lu_3Os_4Ge}_{12.53}$ (Fig. S2c) R_H is negative, indicating electron-*like* charge carrier only. The resulting carrier density n at T = 2 K is $n \approx 10^{19}$ - $10^{21}\mathrm{cm^{-3}}$, suggesting low carrier densities in all four compounds. For the present 3-4-13 compounds, n is about two to three orders of magnitude smaller than that for good metal superconductors, *e.g.* $\mathrm{Sr_3Ir_4Sn_{13}}$,[6] but in-between the values for poor metals and highly doped semiconductors.[7] As the temperature decreases, a continuous loss of charge carriers is observed for all compounds, as the absolute values $|R_H|$ (not shown) decrease for most of the measured temperature range. The temperature dependence of n is therefore consistent with the resistivity measurements (Fig. 5, main text), since $\rho(T)$ increases as the temperature decreases for all four compounds. Such behavior is also reported in $\mathrm{Y_3Ru_4Ge_{13}}$.[3,8]

The superconducting critical field H_c as a function of temperature is shown in the H-T phase diagrams (Fig. S3). The phase diagram contains T_c and H_c values from specific heat (triangles), resistivity (circles) and magnetization (squares) measurements. The upper critical field $H_{c2}(0)$ is estimated from the H_{c2} fits to $H_{c2}(T) = H_{c2}(0)[(1-t^2)/(1+t^2)]$ with $t = T/T_c$.[9] The resulting $H_{c2}(0)$ values are listed in Table S3. The T = 0 Ginzburg-Landau coherence length $\xi(0)$ can be approximated using the estimated upper critical fields, $H_{c2}(0) = \phi_0/2\pi\ \xi_0^2(0)$ where $\phi_0 = \mathrm{h/2e}$ is the flux quantum. The calculated coherence lengths $\xi_0(0)$ are also listed in Table S3. Similar magnitudes of coherence lengths were reported for the stannide family as well.[9,10,12] The thermodynamic critical field $H_c(0)$ and $\kappa(0)$ can be estimated using the expressions $H_c(0) = 4.23\ {\gamma_n}^{1/2}T_c$ and $\kappa(0) = \frac{H_{c2}(0)}{\sqrt{2}H_c(0)}$,[13,14] and are listed in the Table S3. The $\kappa(0) \sim 700 >> \frac{1}{\sqrt{2}}$ indicates that these are type *II* superconductors.

TABLE S1: Calculated ADP ratios

Compound	$B_{iso/eq}(2a)$	$B_{iso/eq}(T)$	$U_{iso/eq}(2a)$	$U_{iso/eq}(T)$	ADP ratio
$Lu_3Co_4Ge_{13}$	-	-	0.0225	0.0050	4.5
$Lu_3Rh_4Ge_{13}$	-	-	0.0215	0.0037	5.8
$Lu_3Os_4Ge_{12.53}$	-	-	0.0320	0.0048	6.7
$Y_3Os_4Ge_{12.65}$	-	-	0.0400	0.0050	8.0
$Y_3Ir_4Ge_{13}$	-	-	0.0117	0.0009	13.2
$Y_3Rh_4Ge_{12.93}$	-	-	0.0130	0.0008	16.2
$Yb_3Co_4Ge_{13}$	0.65	0.69	0.0082	0.0087	0.9
$Yb_3Co_{4.3}Sn_{12.7}$	1.80	1.39	0.0228	0.0176	1.3
$La_3Co_4Sn_{13}$	-	-	0.0184	0.0113	1.6
$Ce_3Co_4Sn_{13}$	-	-	0.0151	0.0085	1.8
$Yb_3Co_4Sn_{13}$	-	-	0.0161	0.0064	2.5
$Yb_3(Co,Ru)_4Ge_{13}$	-	-	0.0137	0.0050	2.7

TABLE S2: Selected interatomic distances in $Lu_3T_4Ge_{13-x}$ (T = Co, Rh, Os) and $Y_3Os_4Ge_{13-x}$.

	$Lu_3Co_4Ge_{13}$	**$Lu_3Rh_4Ge_{13}$**	**$Lu_3Os_4Ge_{12.53}$**	**$Y_3Os_4Ge_{12.65}$**
Ge icosahedron				
Ge1-Ge2A (x12)	2.6615(54)	2.6411(40)	3.0448(113)	3.0921(8)
Ge1-Ge2B (x12)	3.1484(31)	3.2641(24)	3.4823(556)	-
R cuboctahedron				
R-Ge2A (x8)	3.2047(45)	3.2493(37)	3.1097(95)	3.1337(12)
R-Ge2A (x4)	3.1100(61)	3.2634(44)	3.2015(141)	3.1910(15)
R-Ge2B (x8)	2.9931(27)	2.9966(21)	3.0740(504)	-
R-Ge2B (x4)	3.0377(43)	3.1451(35)	2.9950(771)	-
T trigonal prism				
T-Ge2A (x6)	2.3721(61)	2.4544(46)	2.4727(56)	2.4790(17)
T-Ge2B (x6)	2.4050(35)	2.4839(27)	2.4766(322)	-

TABLE S3: Physical properties parameters of $Lu_3T_4Ge_{13-x}$ (T = Co, Rh, Os) and $Y_3Os_4Ge_{12.7}$

R	T	T_c (K)	γ_n (mJ$mol^{-1}K^{-2}$)	β (mJ$mol^{-1}K^{-4}$)	DOS(E_F) (states eV^{-1} f.u.$^{-1}$)	γ_{DFT} (mJ$mol^{-1}K^{-2}$)	θ_D (K)	$\frac{\Delta C_e}{\gamma_n T_c}$	$\frac{2\Delta(0)}{k_B T_c}$	$H_{c2}(0)$ (T)	$\xi(0)$ (nm)	$H_c(0)$ (Oe)	$\kappa(0)$
Lu	Co	1.40 (3)	5.35	0.301	6.70	5.25	506	1.02	3.12	2.10(6)	15.3	13.80	718
Lu	Rh	1.41(3)	5.19	0.299	6.40	5.02	505	1.07	2.95	1.98	12.9	20.23	692
Lu	Os	2.51(1)	8.55	0.406	7.17	5.62	537	1.48	2.96	3.20	10.1	31.04	729
Y	Os	2.03(3)	8.25	0.566	9.38	7.35	589	1.30	3.02	2.70	11.1	24.66	744

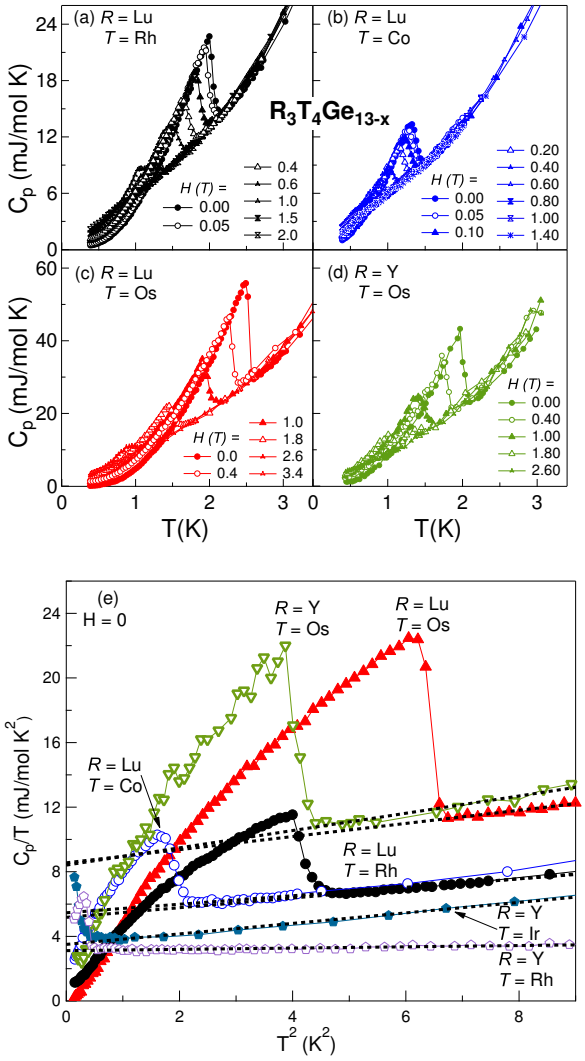

FIG. S1: Temperature-dependent specific heat for (a) R = Lu and T = Rh, (b) R = Lu and T = Co, (c) R = Lu and T = Os, and (d) R = Y and T = Os. (e) C_p/T *vs.* T^2 plot with dotted lines representing linear fits just above T_c.

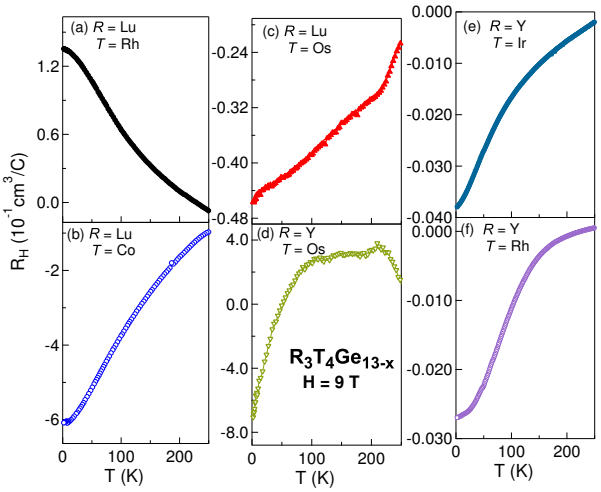

FIG. S2: H = 9 T temperature-dependent (2-300 K) Hall coefficient R_H for $R_3T_4Ge_{13-x}$, with (a) R = Lu and T = Rh, (b) R = Lu and T = Co, (c) R = Lu and T = Os, (d) R = Y and T = Os, (e) R = Y and T = Ir, and (f) R = Y and T = Rh.

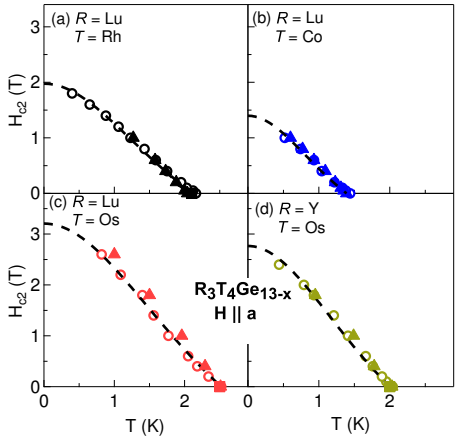

FIG. S3: The $H-T$ phase diagram of $Lu_3T_4Ge_{13-x}$ (T = Co, Rh, Os) and $Y_3Os_4Ge_{12.53}$. The dashed line is a fit to $H_{c2}(T) = H_{c2}(0)[(1-t^2)/(1+t^2)]$ with $t = T/T_c$. The points were determined from $C_p(T)$ (triangles), $\rho(T)$ (circles), and M(T) (squares).

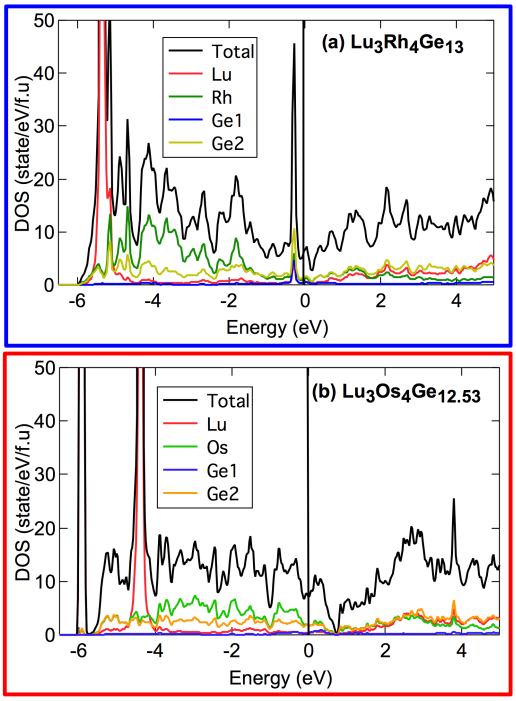

FIG. S4: DOS plots for (a) $Lu_3Rh_4Ge_{13}$ without spin-orbit coupling (SOC) (b) $Lu_3Os_4Ge_{12.53}$ with SOC. Total and partial DOS of Lu, T, Ge1 and Ge2 are black, red, green, blue, and orange, respectively. Fermi levels are marked by vertical lines.

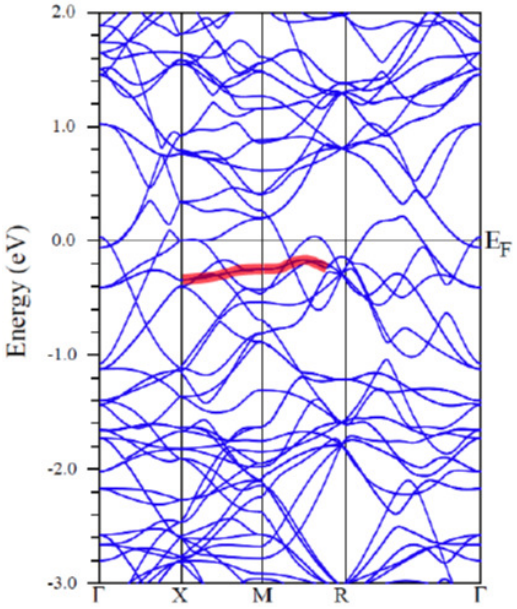

FIG. S5: Band structure plot for $Lu_3Rh_4Ge_{13}$ with a flat band between -0.4 to -0.2 eV highlighted with a thick red curve.

III. REFERENCES

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