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CRYSTAL GROWTH AND THE SEARCH FOR HIGHLY CORRELATED INTERMETALLICS

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CRYSTAL GROWTH AND THE SEARCH FOR HIGHLY CORRELATED INTERMETALLICS

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Our interests lie in studying and understanding the structure-property relationships in novel ternary intermetallics synthesized by metallic flux-growth methods. This review highlights the synthesis, structures and physical properties of ternary lanthanide – transition metal – main group element intermetallics ($Ln - T - X$), where $T =$ Co, Rh, Ir, Ni, Pd, and Pt, and $X =$ Ga, In, Sb and Sn. Several phases will be discussed, including $Ln_nT\text{In}_{3n+2}$ ($n = 1,\ 2,\ \infty$), CePdGa_6, $\text{Ce}_2\text{PdGa}_{12}$, $\text{Ce}_2\text{PdGa}_{10}$, $Ln\text{NiSb}_3$, $Ln\text{Ni}_{1-x}\text{Sb}_2$ ($x \sim 0.4$) and $Ln_3\text{Co}_4\text{Sn}_{13}$. In addition, we report the synthesis, structure characterization, and magnetic behavior from single crystals of $Ln_3\text{Co}_4\text{Sn}_{13}$ ($Ln =$ Pr, Nd, Sm, Gd, Tb).

***Keywords*: heavy fermion, superconductivity, magnetism, heat capacity, electrical resistivity, rare earth intermetallics, single crystal, flux growth, indides, gallides, antimonides, stannides, CeCoIn_5, CeRhIn_5, CeIrIn_5, Ce_2CoIn_8, Ce_2RhIn_8, Ce_2IrIn_8, CePdGa_6, $\text{Ce}_2\text{PdGa}_{10}$, $\text{Ce}_2\text{PdGa}_{12}$, CeNiSb_3, LnNiSb_2, YNiSb_2, $\text{Ln}_3\text{Co}_4\text{Sn}_{13}$, $\text{Pr}_3\text{Co}_4\text{Sn}_{13}$**

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INTRODUCTION

Highly correlated electron systems are of considerable interest, particularly in the design, discovery and growth of novel bulk materials. One of the primary driving forces of basic research in the physics of new materials is the synthesis of these materials in single crystalline form. The study of the magnetic and transport behavior of single crystals allows for a fundamental correlation of the structures and physical properties of these materials.

Our interests lie in the crystal growth, chemical structures, and understanding of the physical properties of new rare-earth intermetallics, which may display signature behavior such as superconductivity and/or heavy fermion behavior, enhanced magnetic susceptibility with local-moments at high temperatures, intermediate valence, and large magnetoresistance (MR). Some of the phenomena we seek to address include the interactions between the *f*-electrons and the conduction electrons in heavy fermion systems. In addition, we also want to study the role of transition metals in heavy-electron systems along with the relationship of the magnetism to superconductivity in these materials.

Why Highly Correlated Materials?

We seek to study heavy fermion behavior because of the parallel to the study of superconductivity.[1–7] Heavy fermions are materials that, at low temperatures, possess enhanced interaction between the *f*-electrons, and the conduction electrons, which results in effective masses $\sim$100 times that of a free electron.[2,4] This phenomenon leads to large electronic specific heat or Sommerfeld coefficient (γ) values obtained from heat capacity measurements with $\gamma \geq 100\,\mathrm{mJ\,mol^{-1}\,K^{-2}}$. In addition, heavy fermion materials may order magnetically at low temperatures, and may also display superconductivity.[4] Most heavy fermion compounds are Ce-, Yb-, or U-based, although a Pr-based heavy fermion, $PrOs_4Sb_{12}$, was discovered and found to be superconducting below $T_c = 1.85\,\mathrm{K}$.[8,9] Moreover, $NdOs_4Sb_{12}$ and $SmOs_4Sb_{12}$ are also newly discovered heavy fermion compounds.[10–14] Some of the well-known and recently found heavy fermion systems are listed in Table 1. Examples (those with superconducting transitions are followed by a corresponding T_c value) include $CeCu_2Si_2$, ($T_c \sim 0.50\,\mathrm{K}$),[15] Ce(Co, Rh, Ir)$In_5$ [$T_c \sim 2.3$, 2.2 (17 kbar)[16] and 0.4 K, respectively)],[17,18] YbTCu$_4$ (T = Zn, Ag, Cd, Au),[19] UBe_{13} ($T_c \sim 0.85\,\mathrm{K}$),[20] UPt_3 ($T_c \sim 0.50\,\mathrm{K}$),[21]

Table 1. Examples of heavy-electron systems grouped by structure type with physical properties[a,*]

Compound	Structure type	Ordering, T_C, T_N (K)	γ (mJ mol^{-1} K^{-2})	T_c (K)	Reference
$CeIn_3$	$AuCu_3$	AFM, 11	120[d]	0.175[g]	[53,54,94–97]
Ce_3In	$AuCu_3$	1.8	680[e]	[f]	[98–100]
$CePb_3$	$AuCu_3$	AFM, 1.1	200–250[d]	0.48[g]	[101–105]
$CeAuAl_3$	$BaAl_4$	AFM, 1.32	227[d]	[f]	[106]
$CeCoGe_3$	$BaNiSn_3$	AFM, *complex*	111[d]	[f]	[107,108]
$CeFeGe_3$	$BaNiSn_3$	[b]	150	[f]	[109,110]
$CeIrSi_3$	$BaNiSn_3$	AFM, 5.0	125[e]	[f]	[111,112]
$CeRhSi_3$	$BaNiSn_3$	AFM, 1.8	120[e]	[g]	[111–113]
$CeCu_2Sb_2$	$CaBe_2Ge_2$				[114,115]
$CeNi_2Sb_2$	$CaBe_2Ge_2$/ $ThCr_2Si_2$	AFM, (1)	400[e]	[f]	[115,116]
$CePdSb_3$	$CaBe_2Ge_2$	[b]	250[e]	[f]	[116]
$CeCu_2Sn_2$	$CaBe_2Ge_2$	AFM, 1.6		[f]	[117]
$CeIr_2Sn_2$	$CaBe_2Ge_2$	AFM, 4.1		[f]	[117]
$CeNi_2Sn_2$	$CaBe_2Ge_2$	AFM, 1.8	600[e]	[f]	[117–120]
$CePd_2Sn_2$	$CaBe_2Ge_2$	AFM, 0.50		[f]	[117,121]
$CePt_2Sn_2$	$CaBe_2Ge_2$	AFM, 0.88	3500[e]	[f]	[117,122]
$CeRh_2Sn_2$	$CaBe_2Ge_2$	AFM, 0.47		[f]	[117]
$Ce_{24}Co_{11}$	$Ce_{24}Co_{11}$	[b]	1800[d]	[f]	[123–126]
$CeCu_2$	$CeCu_2$	AFM, 3.5	82–90[d]	[f]	[127,128]
$Ce(Cu_xGa_{1-x})_2$	$CeCu_2$	(AFM → FM)	115	[f]	[128]
CeCuGa	$CeCu_2$	[c]	110	[f]	[128–130]
$CeCu_4Al$	$CeCu_5$	[b]	280[d], 2000[e, high field]	[f]	[131–133]
$CeCu_4Ga$	$CeCu_5$	[b]	3150[e]	[f]	[134,135]
$CeCu_2Zn_2Al$	$CeCu_5$	[b]	1800[e, high field]	[f]	[132,133]
$CeCu_6$	$CeCu_6$	[b]	1300–1600[e]	[f]	[136–140]
$Ce(Cu_{0.9}Ag_{0.1})_6$	$CeCu_6$	[b]	2800[e]	[f]	[141]
CeRuSi	CeFeSi	[b]	220[e]	[f]	[142,143]
$CeInCu_2$	$CeInCu_2$	[b]	1200[e]	[f]	[144–146]
$Ce_5Ni_6In_{11}$	$Ce_5Ni_6In_{11}$	AFM, 0.63 & 1.10	145[d]	[f]	[147]
$CeCo_{0.89}Ge_2$	$CeNiSi_2$	[b]	128[d]	[f]	[148]
$CeNiGe_2$	$CeNiSi_2$	AFM, 3.9 & 3.2	98[d]	[f]	[149–151]
$CeNi_9Ge_4$	$CeNi_9Ge_4$	[b]	5500[e]	[f]	[152,153]
$CeNi_9Si_4$	$CeNi_9Si_4$/ $NaZn_{13}$	[c]	155[e]	[f]	[154–156]
$Ce_5Ni_2Si_3$	Ce_2NiSi	AFM, 7.3	600[e]	[f]	157
$CePdGa_6$	$CePdGa_6$	AFM, 5.5	230–400[d]	[f]	[41,56]
$Ce_8Pd_{24}Bi$	$Ce_8Pd_{24}Sb$	AFM, 4.6	1360[d]/1400[e]	[f]	[158]
$Ce_8Pd_{24}Ga$	$Ce_8Pd_{24}Sb$	AFM, 3.1	3300[e]	[f]	[158]

(*Continued*)

Table 1. Continued

Compound	Structure type	Ordering, T_C, T_N (K)	γ (mJ mol^{-1} K^{-2})	T_c (K)	Reference
$Ce_8Pd_{24}In$	$Ce_8Pd_{24}Sb$	AFM, 2.5	1720[d]/1000[e]	[f]	[158,159]
$Ce_8Pd_{24}Pb$	$Ce_8Pd_{24}Sb$	AFM, 5.8	1160[d]/790[e]	[f]	[158]
$Ce_8Pd_{24}Sb$	$Ce_8Pd_{24}Sb$	AFM, 6.4 & 5.1	400[e]	[f]	[158,160]
$Ce_8Pd_{24}Sn$	$Ce_8Pd_{24}Sb$	AFM, 5.7	1030[d]/1900[e]	[f]	[158]
$CePt_3Si$	$CePt_3B$	AFM, 2.2	390[e]	0.75	[38]
$CeRuGe_3$	$CeRuGe_3$	(FM)	107[d]	[c]	[161]
CePdIn	Fe_2P	AFM, 1.8	700[e]	[f]	[162–164]
CePtIn	Fe_2P	[b]	500[e]	[f]	[162,165,166]
$Ce_3Cu_4Ge_4$	$Gd_3Cu_4Ge_4$	AFM, 10.3 7.8, 2.6	225[d]	[f]	[167]
$Ce_3Cu_4Sn_4$	$Gd_3Cu_4Ge_4$	AFM, 10.3, 9 7.3, 2.6	330[d]	[f]	[167,168]
$CeCoIn_5$	$HfCoGa_5$	[b]	290[e]	2.3	[18]
$CeIrIn_5$	$HfCoGa_5$	[b]	750[e]	0.4	[17]
$CeRhIn_5$	$HfCoGa_5$	AFM, 3.8	400[e]	2.1[g]	[16]
Ce_2IrIn_8	Hf_2CoGa_8	[b]	700[e]	[f]	[52]
Ce_2RhIn_8	Hf_2CoGa_8	AFM, 2.8	400[e]	2.0[g]	[51]
Ce_6Ni_2Sn	Ho_6Ni_2Ga	[b]	160[e]	[f]	[169]
Ce_3Al_{11}	La_3Al_{11}	FM, 6.2; AFM, 3.2	120	[f]	[170–173]
$Ce_{0.9}Fe_3CoSb_{12}$	$LaFe_4P_{12}$	[b]	350[d]	[f]	[174]
$CeFe_4Sb_{12}$	$LaFe_4P_{12}$	[b]	180[d]	[f]	[174]
$Ce(Pt_{1-x}Au_x)Si$	LaPtSi	[b]	[c]	[f]	175
CePtSi	LaPtSi	[b]	800[e]	[f]	[176]
$CeAl_2$	$MgCu_2$	AFM, 3.8	135[e]–150	[f]	[177–182]
$CePb_2$	$MoSi_2$	AFM, 3.6	200[d]	[f]	[183]
$CeAl_3$	Ni_3Sn	[b]	1620[e]	[f]	[184]
Ce_3Al	$Ni_3Sn/\alpha,\gamma$-Ce_3Al	AFM, 2.5	200[d]		[185]
$CePd_2Al_3$	$PrNi_2Al_3$	AFM, 2.8	380[e]	[f]	[186]
$CeNiGa_3$	$ThCr_2Si_2/CePtGa_3$	AFM, [c]	250[e]	[f]	[116]
$CeCu_2Ge_2$	$ThCr_2Si_2$	AFM, 4.15		0.64[g]	[187–189]
$CeNi_2Ge_2$	$ThCr_2Si_2$	MM	350[e]	0.22[g]	[190–193]
$CeCu_2Si_2$	$ThCr_2Si_2$	AFM, 0.7	1100[e]	0.65	[194]
$CePd_2Si_2$	$ThCr_2Si_2$	AFM, 10	250	0.4[g]	[54,195]
$CeRh_2Si_2$	$ThCr_2Si_2$	AFM, 39 & 27		400 mK[g]	[195]
$CeRu_2Si_2$	$ThCr_2Si_2$	AFM, 1.5	385[e]	[f]	[196]
Ce_7Ni_3	Th_7Ni_3	AFM, 1.8	1200[d]/ 9000[e, high field]	[f]	[197–199]
CeNiSn	ε-TiNiSi	(AFM)	190–200	[f]	[200,201]
$Ce_2Cu_8Al_9$	Th_2Zn_{17}	[b]	1900[e]	[f]	[202]
$Ce_2Ir_3Ge_5$	$U_2Co_3Si_5$	AFM, 10	152[d]	[f]	[203,204]
$Ce_2Ni_3Ge_5$	$U_2Co_3Si_5$	AFM, 5.1 & 4.5	90[d]	[f]	[205]

(*Continued*)

Table 1. Continued

Compound	Structure type	Ordering, T_C, T_N (K)	γ (mJ mol^{-1} K^{-2})	T_c (K)	Reference
$Ce_2Rh_3Ge_5$	$U_2Co_3Si_5$	AFM, 5.5	148[d]	[f]	[203–205]
Ce_5Sn_3	W_5Si_3	(AFM, 17.5)	400[d]	[f]	[206]
$CeNiAl_4$	$YNiAl_4$	[b]	175[e]	[f]	[207,208]
$Ce_3Co_4Sn_{13}$	$Yb_3Rh_4Sn_{13}$	[b]	4000–4200[e]	[f]	[90,209]
$Ce_3Ir_4Sn_{13}$	$Yb_3Rh_4Sn_{13}$	AFM, 0.6	670[e]	[f]	[88,89]
$Ce_3Pt_4In_{13}$	$Yb_3Rh_4Sn_{13}$	AFM, 0.95	1000[e]	[f]	[87]
$Ce_3Ru_4Ge_{13}$	$Yb_3Rh_4Sn_{13}$	(FM)	111[d]	[f]	[161]
$Ce_{4-x}Ru_4Ge_{12+x}$	$Yb_3Rh_4Sn_{13}$		492–592	[f]	[161]
$CeRuSn_3$	(Yb)$Pr_3Rh_4Sn_{13}$	(AFM), 0.6	1670[e]	[f]	[210,211]
$PrInAg_2$	BiF_3	[b]	6500[e]	[f]	[212]
$PrFe_4P_{12}$	$LaFe_4P_{12}$	AFM, 6.4	1000[e]	[f]	[213–215]
$PrOs_4Sb_{12}$	$LaFe_4P_{12}$	[c]	600[d]	1.85	[216, 217]
$NdOs_4Sb_{12}$	$LaFe_4P_{12}$	FM, 1	435–530[d]	[f]	[10–12]
$SmFe_4Sb_{12}$	$LaFe_4P_{12}$	FM, 1.6	370[e]	[f]	[218–220]
$SmOs_4Sb_{12}$	$LaFe_4P_{12}$	FM, 2.6	880[d]	[f]	[12–14]
$YbCu_{5-x}Ag_x$	$AuBe_5/CaCu_5$	[c]	210–460[d]	[f]	[221]
$YbAgCu_4$	$AuBe_5$	[b]	245[d]	[f]	[222]
$YbAuCu_4$	$AuBe_5$	<1	150[d]	[f]	[19,222]
$YbCdCu_4$	$AuBe_5$	[b]	174[d]	[f]	[19]
$YbPdCu_4$	$AuBe_5$	<1	200[d]	[f]	[19]
$YbZnCu_4$	$AuBe_5$	[b]	230[d]	[f]	[19]
YbSi	CrB	AFM, 1.6	600	[f]	[223,224]
YbPd	CsCl	AFM, [c]	600[=]	[f]	[225]
YbCuAl	Fe_2P	(AFM), 28	260[d]	[f]	[226,227]
$YbNi_2B_2C$	$LuNi_2B_2C$	[b]	200[d]/530[e]	[f]	[228,229]
YbPdBi	MgAgAs	[b]	470[d]	[f]	[230]
YbBiPt	MgAgAs	(AFM), <1	8000[e]	[f]	[27,231]
YbNiSb	MgAgAs	[b]	150–175[d]	[f]	[232,233]
YbPdSb	MgAgAs	AFM, <1	240[d]	[f]	[230,234]
Yb_2Ni_2Al	$Mo_2Ni(B/Al)_2$	[b]	700[e]	[f]	[235]
$YbCu_2Si_2$	$ThCr_2Si_2$	[g]	135	[f]	[236,237]
$YbIr_2Si_2$	$ThCr_2Si_2/CaBe_2Ge_2$	[b]	370[e]	[f]	[238]
$YbPd_2Si_2$	$ThCr_2Si_2/BaAl_4$	[b]	203	[f]	[239–241]
$YbRh_2Si_2$	$ThCr_2Si_2$	AFM, 65 mK	370[e]	[f]	[238,242]
Yb_4As_3	anti-Th_3P_4	[b]	205[d]	[f]	[243]
YbPtAl	ε-TiNiSi	AFM, 5.8	200	[f]	[244,245]
YbNiGa	ε-TiNiSi	[c]	450[d]	[f]	[246]
YbRhSb	ε-TiNiSi	FM, 2.7	188[d]	[f]	[247]
YbNiSn	ε-TiNiSi	FM, 5.6	300	[f]	[248]
$Yb_2Co_3Ga_9$	$Y_2Co_3Ga_9$	[b]	112[d]	[f]	[249]
$YbCu_{3.5}$	($YbCu_{3.5}$)	[c]	270[d]	[c]	[250]
$YbCu_{4.5}$	$YbCu_{4.5}$	[b]	430[d]/560[e]	[f]	[250–252]

(*Continued*)

Table 1. Continued

Compound	Structure type	Ordering, T_C, T_N (K)	γ (mJ mol^{-1} K^{-2})	T_c (K)	Reference
YbNiAl	ZiNiAl	AFM, 3	350[d]	[f]	[244,253]
YbAgGe	ZrNiAl	<1	570[e]	[f]	[254–256]
YbPtIn	ZrNiAl	(AFM), 3.5	430[d]/700[e]	[f]	[246,257]
YbPtSn	ZrNiAl	AFM, 3.5	370[e]	[f]	[234,258,259]
YbRhSn	ZrNiAl	AFM, 2	360[d]/1200[e]	[f]	[234,246,260]
U_2PdSi_3	AlB_2	(FM), 75	180[e]	[f]	[261,262]
U_2PtSi_3	AlB_2	FM, (90)	200[d]		[263,264]
U_3Si_5	AlB_2	[b]	110[d]	[f]	[264–266]
$UAgCu_4$	$AuBe_5$	AFM, 18	310	[f]	[267,268]
UCu_5	$AuBe_5$	AFM, 16	86[d]	[f]	[267]
UCu_4Pd	$AuBe_5$	[c]		[f]	[269–271]
$UCu_{3.5}Pd_{1.5}$	$AuBe_5$	[b]		[f]	[269,270]
$UAuPt_4$	$AuBe_5$	[b]	725[e]	[f]	[272]
USn_3	$AuCu_3$	[b]	171[d]	[f]	[273,274]
UPd_2Ga_3	BaB_2Pt_3	AFM, 13	230[d]	[f]	[275]
UCd_{11}	$BaHg_{11}$	AFM, 5	840[d]	[f]	[276]
UIr_2Si_2	$CaBe_2Ge_2$	AFM, 6	105[d]/300[e]	[f]	[277,278]
UCu_5In	$CeCu_5Al$	AFM, 23	170[d]	[f]	[279]
UCu_5Sn	$CeNi_5Sn$	Ferri-, 53.5	330[d]	[f]	[279–281]
UPdIn	Fe_2P	AFM, 21	280[e]	[f]	[282–285]
$U_{14}Au_{51}$	$Gd_{14}Ag_{51}$	AFM, 22	315[d]	[f]	[286,287]
$U_3Ni_5Al_{19}$	$Gd_3Ni_5Al_{19}$	AFM, 23	185[e]	[f]	[288]
UAl_2	$MgCu_2$	[b]	142[e]	[f]	[289–291]
UBe_{13}	$NaZn_{13}$	AFM, 8.8	1100[e]	0.85	[20,292,293]
UPt_3	Ni_3Sn	AFM, 5.5	450[d]	0.5	[21,294,295]
UNi_2Al_3	$PrNi_2Al_3$	AFM, 4.6	120[e]	1	[24]
UPd_2Al_3	$PrNi_2Al_3$	AFM, 14	140–150[e]	2	[25,296]
$UPd_2(Al_{1-x}Ga_x)_3$	$PrNi_2Al_3/BaB_2Pt_3$	AFM, ~10–14.5	~150–230[e]	~0.33–1.9	[297]
URh_2Ge_2	$ThCr_2Si_2/CaBe_2Ge_2$	[b]	130[d]	[f]	[298,299]
URu_2Si_2	$ThCr_2Si_2$	AFM, 17.5	75–180[d]	1.5	[300–302]
U_2Zn_{17}	Th_2Zn_{17}	AFM, 9.7	500[d]	[f]	[303]
UCu_5Al	UCu_5Al	AFM, 18	180[d]	[f]	[279,304]
U_2PtC_2	U_2IrC_2	[b]	75[d]	1.47	[305]
U_2RuSi_3	U_2RuSi_3	[b]	115		[261–306]
U_2Ni_2In	U_3Si_2	AFM, 15	200[e]	[f]	[307,308]
U_2Pd_2In	U_3Si_2	AFM, 38	393[e]	[f]	[307,309]
U_2Rh_2In	U_3Si_2	[b]	280[e]	[f]	[307,308]
U_2Ir_2Sn	U_3Si_2	[b]	130[e]	[f]	[307]
U_2Ni_2Sn	U_3Si_2	AFM, 25		[f]	[307]
U_2Pd_2Sn	U_3Si_2	AFM, 41	203[e]	[f]	[307,309]
U_2Pt_2Sn	U_3Si_2	AFM, 15.5	334[e]	[f]	[307]
U_2Rh_2Sn	U_3Si_2	AFM, 24	131[e]	[f]	[307]

(*Continued*)

Table 1. Continued

Compound	Structure type	Ordering, T_C, T_N (K)	γ (mJ mol^{-1} K^{-2})	T_c (K)	Reference
$U_3Au_3Sn_4$	$Y_3Au_3Sb_4$	[b]	280[e]	[f]	[310]
$U_3Cu_3Sn_4$	$Y_3Au_3Sb_4$	AFM, 12	380[e]	[f]	[310]
$U_3Ni_3Sn_4$	$Y_3Au_3Sb_4$	[b]	92[e]	[f]	[310]
$U_3Pt_3Sn_4$	$Y_3Au_3Sb_4$	[b]	94[e]	[f]	[310]
U_2Pt_2In	Zr_3Al_2/U_3Si_2	[b]	830[e]	[f]	[307,311–313]
$NpSn_3$	$AuCu_3$	AFM, 9.5	242[e]	[f]	[314,315]
$NpBe_{13}$	$NaZn_{13}$	AFM, 3.4	900[e]	[f]	[292,316]
$PuCoGa_5$	$HoCoGa_5$	DM, 18.5	77[e]	18.5	[317]

[a]Abbreviations: T_C, Curie temperature; T_N, Néel temperature; γ, Sommerfeld coefficient; T_c, superconducting transition (critical) temperature; AFM, antiferromagnetic; FM, ferromagnetic; MM, metamagnetic; Ferri-, ferrimagnetic; DM, diamagnetic.

[b]Pauli, Curie-Weiss or Van Vleck paramagnet, or does not magnetically order down to lowest temperature measured, or was not measured/reported.

[c]not explicitly stated/reported.

[d]C_p/T or C_m/T at ambient pressure, or the magnetic contribution to the specific heat determined by subtracting out the lattice (phonon) contribution β and plotted versus T^2; the linear specific heat coefficient (γ) is determined by extrapolating data above phase transition to $T = 0$ K and estimated from least-squares fits to equation $C_p/T = \gamma + \beta T^2$ [where the Debye phonon contribution $\beta = 12\pi^4 rR/(5\Theta_D^3)$, r is the number of atoms per formula unit, R is the universal gas constant, and Θ_D is the Debye temperature].

[e]low-temperature specific heat at phase transition (kink), or maximum in C/T, or estimate of C/T vs T data extrapolated to $T = 0$ K, or C/T at lowest temperature.

[f]not measured/reported or is not superconducting.

[g]substance undergoes magnetic transition or becomes superconducting under applied pressure.

*For articles referenced in this table see also references therein. Data collected using samples from various studies may not agree due to variations within the samples such as phase purity, stoichiometry and lattice disorder, and may also be due to errors made during measurements. Parentheses indicate possible magnetic ordering of the specified type.

URu_2Si_2 ($T_c \sim 1.5$ K),[22,23] UNi_2Al_3 ($T_c \sim 1.0$ K),[24] and UPd_2Al_3 ($T_c \sim 2.0$ K).[25,26] YbBiPt has $\gamma \sim 8000$ mJ mol^{-1} K^{-2}, which to the best of our knowledge is the highest value for any heavy fermion material.[27] We note that there are a variety of structure types that these compounds adopt. They include the well-known tetragonal $ThCr_2Si_2$[28] (with >700 compounds crystallizing with this structure),[29] tetragonal $CaBe_2Ge_2$,[30] hexagonal Ni_3Sn,[31] hexagonal ZrNiAl,[32] hexagonal $PrNi_2Al_3$,[33] cubic $AuBe_5$,[34] and cubic $NaZn_{13}$[35] structure types.

Our search for highly correlated systems has in part been motivated by the search for materials for spintronics, a new technology that merges electronics with the manipulation of conduction electron spins. One of the most successful strategies for producing technologically relevant magnetoresistive materials is to enhance the effects of field dependent magnetic scattering processes through the creation of magnetic superlattices,[36,37] or by doping magnetic insulators such that a magnetic and metal–insulator transition coincide. The study of these materials can perhaps lead to the exploitation of "spin" of the electron rather than its charge. An example of a spintronic device is the giant magnetoresistive spin-valve head for magnetic hard-disk drives. Some of the physics that we wish to study include the change of resistivity with magnetic field and temperature. One way to change the properties of a material is to carefully tune the behavior by altering its chemistry (*e.g.,* by elemental substitutions or by doping).

A partial, yet fairly comprehensive, list of heavy fermion systems is provided here, hence our challenge: to identify structural features that favor heavy fermion behavior. We begin to see that compounds with high symmetry are favorable for heavy fermion superconductivity; however, we must now also consider the role of local symmetry in compounds such as noncentrosymmetric $CePt_3Si$.[38] To correlate the structures and the physical behavior of these materials, some other characteristics to consider include rare earth environments, lattice dimensionality, and structural units. Our approach to such a challenge is considered both exploratory and systematic.

In this review, we will highlight several of the ternary intermetallic systems that we have investigated, each adopting one of the following structure types: $HoCoGa_5$,[39,40] Ho_2CoGa_8,[39,40] $CePdGa_6$,[41] $CeNiSb_3$,[42] $HfCuSi_2$[43] or $Yb_3Rh_4Sn_{13}$.[44] We begin by describing the synthetic methods and characterization techniques, followed by discussions that summarize the studies of the indium-, gallium-, and antimony-containing compounds. Herein, we also report the synthesis and full characterization of the structure and magnetic properties from single crystals of $Ln_3Co_4Sn_{13}$ (Ln = Pr, Nd, Sm, Gd–Er). Our goal is to present new systems in which the phenomena and/or implications of heavy fermion behavior, superconductivity, magnetism and/or magnetoresistance may be observed and studied, and the problem of correlating these characteristics with structural features may be addressed.

SYNTHESIS AND CHARACTERIZATION

Flux Growth

In the synthesis of many *new* ternary intermetallic phases, we have explored the use of several main group elements as metallic fluxes. The flux growth method has been employed as a synthetic route to forming many compounds—mostly binaries of the transition metal borides, carbides and silicides.[45] Further details on the various flux-growth methods have been described elsewhere.[46,47] This technique has afforded us the opportunity to grow several novel and interesting phases, and also phases previously synthesized and characterized by polycrystalline methods. The relatively low melting points of the main group elements allow for phase formation at low and manageable temperatures. These metallic fluxes serve as a solvent and in most cases are incorporated into the structure. In this section, we discuss the specifics for crystal growth, structure characterization and physical property measurements used in our studies. Detailed synthesis and characterization information for each ternary system will be provided in subsequent sections.

Our sample preparation begins by first evaluating the binary phase diagrams within each intermetallic ternary system. This, when taken into account with the melting points of the flux elements (gallium, indium, tin or antimony), serves as a precursor to generating a carefully designed temperature profile. The profile is designed to prevent secondary phases from forming in the melt, to allow for congruent melting of the starting materials, and ultimately to stimulate the rapid growth of large single

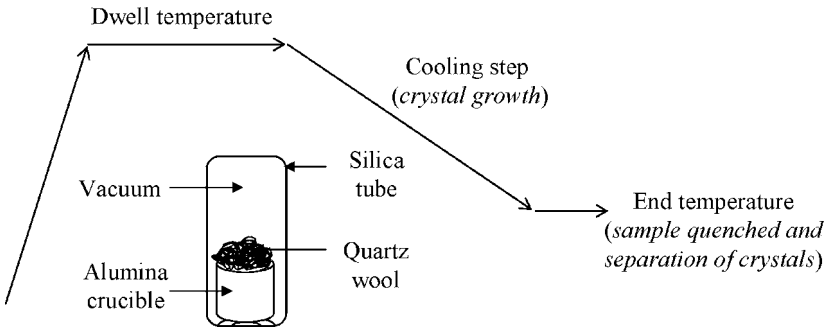

Figure 1. Shown is a schematic diagram of the flux-growth reaction vessel with a simple temperature profile.

Figure 2. A selection of single crystals from several of our growths.

crystals. Samples are made by first combining the elements in precisely calculated ratios into alumina crucibles, which are covered with quartz wool then sealed into evacuated quartz tubes. The air-free environment inside the fused silica ampoule prevents oxidation from occurring during crystal growth. Figure 1 shows a diagram of the reaction vessel with a simple temperature profile. After reaching the appropriate temperature at the end of a temperature cycle (always above the melting point of the flux metal), the excess flux, or solvent, is still liquidous and is filtered through the quartz wool by centrifugation. The single crystals are left behind and are mechanically extracted, after which the remaining flux is chemically etched from the surfaces whenever necessary, and properly stored for characterization. A selection of single crystals from several of our growths is shown in Figure 2.

X-ray Diffraction

Structure determination is performed using X-ray diffraction (XRD). Powder XRD (using a Bruker D-8 X-ray Diffractometer with monochromatized Cu K_α radiation, $\lambda = 1.540562\,\text{Å}$) is used to determine the homogeneity of each sample batch by collecting data on ground single crystals.

Single crystal XRD (using a Nonius Kappa CCD X-ray Diffractometer with graphite monochromatized Mo K_α radiation, $\lambda = 0.71073$ Å) is also used to confirm the sample homogeneity by performing multiple lattice determinations on several single crystals. Direct methods are employed to solve the structures using the SHELXL97 software package.[48]

Physical Property Measurements

Magnetic properties are measured on single crystals or aggregates of single crystals using a Quantum Design Superconducting Quantum Interface Device (SQUID) magnetometer. Transport measurements are made using a standard four-lead method with a Quantum Design Property Measurement System (PPMS) at ambient pressure. Specific heat data is measured with a Quantum Design PPMS using a thermal relaxation method from ~ 0.36 to ~ 30 K in zero applied field.

Indium: $Ln_nT\text{In}_{3n+2}$ (Ln = La, Ce; T = Co, Rh, Ir ; n = 1, 2, ∞)

The $Ln_nT\text{In}_{3n+2}$ family of compounds, shown in Figure 3(a,b), offers the unique opportunity to closely examine the coexistence of magnetism and superconductivity.[16–18,49,50] The structure(s) consists of Ce cuboctahedra (CeIn_3) and $T\text{In}_2$ layers. The $\text{Ce}T\text{In}_5$ (T = Co, Rh, Ir; $n = 1$ member) compounds each become superconducting at temperatures

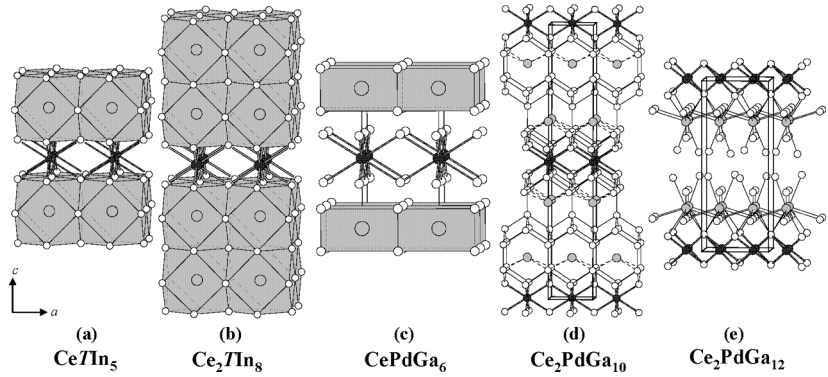

Figure 3. The structures of (a) CeCoIn_5, (b) Ce_2RhIn_8, (c) CePdGa_6, (d) $\text{Ce}_2\text{PdGa}_{10}$ and (e) $\text{Ce}_2\text{PdGa}_{12}$ are shown for comparison with unit cells outlined as solid lines. The light grey circles are Ce atoms, the black circles are the T elements (Co, Rh, Ni), and the white circles are the X elements (In, Ga).

below ca. 2.3 K: $CeCoIn_5$ and $CeIrIn_5$ are superconducting at 2.3 and 0.4 K, respectively, while $CeRhIn_5$ superconducts at 2.1 K under applied pressures of 16 kbar.[16–18]

At ambient pressure, $CeRhIn_5$ is a heavy fermion antiferromagnet with an incommensurate magnetic structure and $T_N \sim 3.8$ K.[16] We note that the $n = 2$ analogue, Ce_2RhIn_8, orders antiferromagnetically at $T_N \sim 2.8$ K at ambient pressure, but superconductivity at $T_c \sim 2$ K can be induced with the application of 25 kbar of pressure.[51] Ce_2IrIn_8, on the other hand, remains paramagnetic to lowest temperatures and does not display superconductivity.[52] $CeIn_3$, the "parent" and $n = \infty$ member of Ce_nTIn_{3n+2}, is cubic and undergoes a commensurate antiferromagnetic transition at $T_N \sim 10.23$ K and is also superconducting with 25 kbar of pressure.[53,54]

The structures of the two Rh compounds are most similar. In $CeRhIn_5$ and Ce_2RhIn_8, the *a*- and *c*-axes of the $CeIn_3$ cuboctahedra are equivalent. The cubic structure is reflected in the *Ln*-In2:*Ln*-In1 ratio, and it is only when this ratio is close to unity that the coexistence of magnetic ordering and superconductivity is found. The implications for the heavy fermion ground state for these materials have been reviewed.[50]

Gallium: Ce_2PdGa_{10}, $CePdGa_6$, and Ce_2PdGa_{12}

In our continued search for heavily correlated electron systems, we also want to explore the effects of transition metal and main group element substitution and the influence on the crystal chemistry. The main group elements surrounding the *f*-element can influence the hybridization, and in metallic compounds this can be viewed as the coupling between the atomic *f*-like level and conduction electrons. We have been able to synthesize several layered ternary Ce-Pd-Ga compounds, namely $CePdGa_6$,[41] Ce_2PdGa_{10}[55] and Ce_2PdGa_{12}.[56] $CePdGa_6$ is a heavy fermion with $\gamma \sim 230$ mJ mol^{-1} K^{-2} and exhibits an anisotropic magnetism with $T_N \sim 5.5$ K.[41,56] Ce_2PdGa_{10} also shows enhanced mass behavior, exhibits paramagnetic behavior down to 2 K, and has a large positive $MR > 200\%$ at 2 K and 9 T.[55] Ce_2PdGa_{12} is an antiferromagnetic heavy fermion with $\gamma \sim 170$ mJ mol^{-1} K^{-2} and a magnetic transition at $T_N \sim 11$ K.[56] These Ce-Pd-Ga phases, shown in Figure 3 (c–e), allow us to examine the influence of the Ce environment, dimensionality and layering on the magnetic and transport properties.

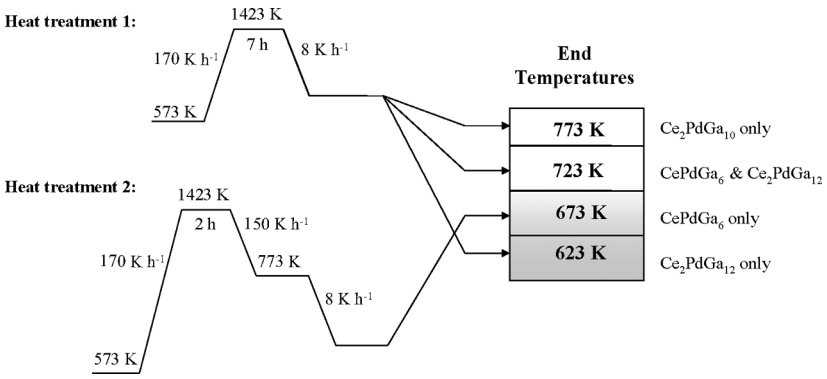

Figure 4. Crystal growth conditions for $CePdGa_6$, Ce_2PdGa_{10} and Ce_2PdGa_{12}.

In order to synthesize phase-pure single crystals of these Ce-Pd-Ga compounds for physical property measurements, it was necessary to first determine optimal crystal growth conditions in which to isolate each phase. To obtain single crystals of homogeneous phases from each growth, we have modified synthetic variables such as the starting stoichiometric ratios and heat treatments. While manipulating these synthetic variables, we found that $CePdGa_6$ and Ce_2PdGa_{12} coexist using identical growth conditions.

Furthermore, we have observed that in addition to modifying the stoichiometric ratios of the starting materials, the heat treatment employed also plays a major role in determining which phase forms. Figure 4 illustrates the growth conditions for these Ce-Pd-Ga phases. We were able to isolate single crystals of the Ce_2PdGa_{12} phase by combining a stoichiometric Ce:Pd:Ga ratio of 1:1:20 and using heat treatment 1.[56] With a modification of the end temperature of heat treatment 1 (773 K), we were able to isolate single crystals of Ce_2PdGa_{10}.[55] However, the synthesis of phase-pure $CePdGa_6$ proved more problematic, requiring not only a change in synthetic variables such as the starting stoichiometric ratios of the reactants, but also in the heat treatment.[56] The successful isolation of $CePdGa_6$ was made possible by combining stoichiometric Ce:Pd:Ga ratios of 1:1.5:15 and following heat treatment 2 (see Figure 4).

The crystal structure of $CePdGa_6$ is layered with a striking resemblance to the Ce_nTIn_{3n+2} (T = Co, Rh, Ir; n = 1, 2, ∞) compounds. Both phases crystallize in the tetragonal $P4/mmm$ (No. 123) space group.

The structure of Ce_nTIn_{3n+2} consists of a periodic stacking of $CeIn_3$ cuboctahedra layers, and $TIn_{8/2}$ rectangular prisms along the *c*-axis. Although exhibiting a similar stacking in the *c*-direction, the structure of $CePdGa_6$ consists of face-sharing $CeGa_{8/4}$ rectangular prisms and $PdGa_{8/2}$ rectangular prisms (shown in Figure 3 c–e for comparison) along the *c*-axis. Ce_2PdGa_{12} crystallizes in the tetragonal $P4/nbm$ (No. 125) space group, and its structure, which shows a similar stacking to that of $CePdGa_6$, can be viewed as alternating $CePdGa_6$ units separated by Ga layers in the *c*-direction. Both structures have Ce-Ga contacts and $PdGa_{8/2}$ rectangular prisms. However, the Ce environments differ in these compounds, where Ce is coordinated to 8 Ga atoms, forming $CeGa_{8/4}$ rectangular prisms in $CePdGa_6$, the Ce environment of Ce_2PdGa_{12} consists of Ce atoms coordinated to 10 Ga atoms. Moreover, Ce_2PdGa_{10} crystallizes in the tetragonal space group $I4/mmm$ (no. 139), and has the Ce-Ga contacts and $PdGa_{8/2}$ rectangular prisms found in $CePdGa_6$ and Ce_2PdGa_{12}. However, the Ce atoms of Ce_2PdGa_{10} are coordinated to 4 Ga atoms.

These layered Ce-Pd-Ga phases allow us to study the effects of layering and rare earth environment on the magnetism in each system. Two competing mechanisms, the RKKY (Ruderman-Kittel-Kasuya-Yosida) and Kondo interactions, can be used to describe the magnetism in these materials.[57–59] RKKY interactions exist when the exchange coupling between the magnetic ion and conduction electrons results in the magnetization of the conduction electrons, which in turn causes an indirect exchange between two distant magnetic ions.[60] We have observed that the higher magnetic ordering in Ce_2PdGa_{12} ($T_N \sim 11$ K) may be attributed to the existence of more Ce-Ga contacts (Ce CN = 10), where the magnetic Ce^{3+} ion can interact with additional conduction electron carriers. These stronger RKKY-like interactions cause the magnetic ordering observed in this compound to be more profound. However, the suppressed magnetism in compounds, such as $CePdGa_6$ ($T_N \sim 5.5$ K) and Ce_2PdGa_{10} (paramagnetic behavior), can be attributed to Kondo-like interactions.[61] The Kondo effect occurs as a result of the shielding of magnetic ions by conduction electrons in magnetic materials. This mechanism has been used to describe a minimum in the electrical resistivity of magnetic materials at a temperature, T_K. In $CePdGa_6$ (Ce CN = 8) and Ce_2PdGa_{10} (Ce CN = 4), there are fewer Ce-Ga contacts, which leads to reduced hybridization, or less interaction between the Ce^{3+} ions and the conduction electrons.[61] By

examining the crystal chemical relationships, we can further examine the two competing effects in these materials.

Antimony: *Ln*NiSb$_3$(Ln = Ce–Nd, Sm) and *Ln*Ni$_{1-x}$Sb$_2$ (*Ln* = Y, Gd–Er; x $\sim$ 0.4)

Antimony serves as a worthy metallic flux element as it lies directly on the metal-insulating border of the periodic table, thus making antimony-containing compounds considerably attractive for their unusual bonding nature and wide variety of magnetic properties.[62–65] The various substructures formed as a result of Sb−Sb bonding within these compounds, such as the formation of Sb^- chains, Sb_2 pairs, Sb_4 squares, and in particular ∞^2[Sb] square nets,[63,64] largely influence the crystal chemistry and also the magnetic and transport properties.[62–65]

We recently discovered a new phase $CeNiSb_3$ while studying the quasi-2D system $CeNiSb_2$ ($HfCuSi_2$-type), which was initially thought to be a heavy fermion with $\gamma \sim 500\,\mathrm{mJ\,mol^{-1}\,K^{-2}}$ (polycrystalline study).[66] Both compounds order ferromagnetically at $T_C \sim 6$ K.[42,67–69] Magnetic data for $CeNiSb_3$ is largely anisotropic with an average effective moment $\mu_{eff} = 2.58\,\mu_B$,[67] which is comparable to the $2.54\,\mu_B$ for the Ce^{3+} free ion. Resistivity data for $CeNiSb_3$ indicate Kondo lattice behavior.[42] Recently published heat capacity data for $CeNiSb_3$ shows a relatively small

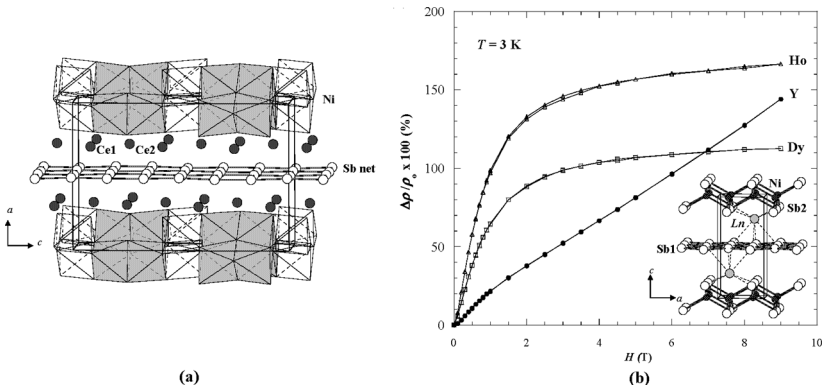

Figure 5. (a) $CeNiSb_3$ structure with unit cell outlined, and black circles representing Ce atoms, white circles representing Sb atoms, and Ni octahedra depicted as open and shaded polyhedra. (b) Magneto-transport ($\Delta\rho/\rho \times 100$) data for $LnNi_{1-x}Sb_2$ (*Ln* = Y, Dy, Ho) compounds at $T = 3$ K. The $LnNi_{1-x}Sb_2$ structure is drawn and labeled in the inset.

Sommerfeld coefficient value $\gamma \sim 50\,\mathrm{mJ\,mol^{-1}\,K^{-2}}$,[70] which is also similar to the $\gamma \sim 60\,\mathrm{mJ\ mol^{-1}\,K^{-2}}$ for *single crystal* $CeNiSb_2$.[71]

The orthorhombic *Pbcm* structure of $CeNiSb_3$ is shown in Figure 5 (a). This highly anisotropic compound has been compared to the structurally similar $CeCrSb_3$,[72] and is composed of layers of buckled and distorted ∞^2[Sb] nets, Ce atoms in mono-capped anti-prismatic geometries, and $\infty^2[NiSb_2]$ octahedra.

To examine the stability of the $CeNiSb_3$ structure type, along with the effects on the structures and physical properties caused by substitutions of the *Ln* element using the rare-earth series Y, Pr, Nd, Sm, and Gd–Yb, we have found that the $CeNiSb_3$ structure only forms for the early lanthanides Pr, Nd, and Sm with our experimental conditions. Yttrium and the mid-lanthanide elements Gd–Er form compounds[73] isostructural to $HfCuSi_2$.[43] However, we note that it may be possible to synthesize the latter lanthanide analogues by other synthetic methods. Similar to $CeNiSb_3$, the $PrNiSb_3$, $NdNiSb_3$, and $SmNiSb_3$ compounds are also highly anisotropic, however each order antiferromagnetically at $T_N \sim 4.5$, 4.6, and 2.9 K, respectively, with effective moments of 3.62, 3.90 and 0.80 μ_B, respectively.[74] The distortions of the substructures in the Sm compound are most pronounced, which suggests severe bond strain within the structure and may be the cause for rendering the latter lanthanide analogues unstable.[73,74]

The structure of the $LnNi_{1-x}Sb_2$ ($x \sim 0.4$) compounds has been described,[73] and is shown in the inset of Figure 5 (b). The structure consists of layers of rare-earth-capped Sb square nets and $NiSb_4$ tetrahedra. By characterizing single crystals of these compounds, we have found that the Ni site is not fully occupied (a common occurrence in many $LnTSb_2$ phases[75–82]) and refinements of the occupancy parameter lead to percentage values close to 60%.[73] The most interesting discovery in this study was the unusually large positive magnetoresistance (*MR*) for the Y, Dy, and Ho compounds $>100\%$ at 9 T. The *MR* data for each $LnNi_{1-x}Sb_2$ compound is shown in Figure 5 (b). The *MR* for the Gd and Tb compounds (not shown) at 9 T is $\sim 8\%$ and 30%, respectively, while the *MR* for the Dy and Ho compounds is $\sim 112\%$ and 170%, respectively. The *MR* for the Y compound approaches 150% with no obvious signs of saturation, a feature reminiscent of $LaSb_2$, which shows linear *MR* up to 45 T, and whose structure contains similar subunits.[83,84] In this study, it was found that although the magnetic ordering temperatures did not scale with the de Gennes factor [$dG = (g-1)^2$

$J(J+1)$],[85] the magnetization and thus the *MR* systematically correlates with the bulk magnetism for each compound.[73] Further measurements are being performed to elucidate the origin of the behavior for the Er compound.

Tin: $Ln_3Co_4Sn_{13}$ (Ln = La–Nd, Sm, Gd, Tb)

Compounds that are isostructural to cubic $Pm\bar{3}n$ $Yb_3Rh_4Sn_{13}$[44] display interesting magnetic and transport properties. $Yb_3Rh_4Sn_{13}$ becomes superconducting below $T_c \sim 8$ K.[44,86] Furthermore, $Er_3Rh_4Sn_{13}$ displays reentrant superconductivity with $T_c \sim 0.97$ K and $T_m \sim 0.57$ K.[86] $Ce_3Pt_4In_{13}$ and $Ce_3Ir_4Sn_{13}$ are heavy fermion compounds with $\gamma \sim 1000$ mJ mol^{-1}K^{-2}[87] and ~ 670 mJ mol^{-1}K^{-2},[88,89] respectively. More recently, the La and Ce compounds of $Ln_3Co_4Sn_{13}$ have been synthesized by Thomas *et al.*[90] and Israel *et al.*[91] $La_3Co_4Sn_{13}$ was found to be a type II superconductor with $T_c \sim 2.4$ K.[90] The Ce-analogue displays heavy fermion behavior with $\gamma \sim 75$ mJ mol^{-1} K^{-2} as estimated from extrapolating C_p/T to 0 K, although C_p/T is enhanced up to 4280 mJ mol^{-1} K^{-2} at the transition peak temperature.[90]

In our study of the *Ln*-Co-Sn system we have been able to synthesize the Pr-, Nd-, Sm-, Gd-, and Tb-analogues of $Ln_3Co_4Sn_{13}$. Single crystals for characterization were grown using lanthanide metals Pr, Nd, Sm, Gd and Tb (99.9%, ingots), Co (99.998%, powder), and Sn (99.8%, shot) purchased from Alfa Aesar. Samples were made using a Sn flux following the aforementioned crystal growth procedures and using the heat treatment: ramp up to 1323 K for 24 h, then slowly cool to 573 K at 5 K hr^{-1}. The irregularly shaped crystals had maximum dimensions of ~ 2 mm^3 and oxidized upon prolonged exposure to air and moisture. Concentrated HCl was used as an etchant.

Initial phase identification of the above samples was determined using powder XRD by indexing peaks from the diffraction pattern of $Yb_3Rh_4Sn_{13}$.[44] The calculated powder patterns from the refined structures were also used to confirm the phase purity of $Ln_3Co_4Sn_{13}$ (Ln = Pr, Pr, Nd, Sm, Gd, Tb). Multiple crystals were selected for characterization by single crystal XRD. Crystal fragments with dimensions of $\sim 0.5 \times 0.5 \times 0.8$ mm^3 were selected for structure determination. Table 2 lists the data collection and refinement parameters for these compounds. The structure of $Pr_3Co_4Sn_{13}$ was solved by first selecting the $Pm\bar{3}n$ space group, and then refined using SHELXL97.[48] The occupancy

Table 2. Data collection and refinement parameters for $Ln_3Co_4Sn_{13}$ (Ln = Pr, Nd, Sm, Gd, Tb)

Compound	Pr	Nd	Sm	Gd	Tb
Space group	$Pm\bar{3}n$	$Pm\bar{3}n$	$Pm\bar{3}n$	$Pm\bar{3}n$	$Pm\bar{3}n$
a(Å)	9.582(6)	9.583(8)	9.5350(6)	9.5190(4)	9.5010(6)
V(Å^3)	879.77(10)	880.0(12)	866.89(9)	862.53(6)	857.65(9)
Z	2	2	2	2	2
Size, min/mid/max (mm^3)	0.5/0.5/0.8	0.5/0.5/0.8	0.5/0.5/0.75	0.5/0.5/0.75	0.5/0.5/0.8
Temperature (K)	298(2)	298(2)	298(2)	298(2)	298(2)
ρ(g cm^{-3})	8.310	8.345	8.542	8.665	10.036
θ range(°)	3.01–30.02	3.01–30.02	3.02–30.00	3.01–29.16	3.03–30.03
μ(mm^{-1})	29.819	30.355	31.993	33.476	33.489
Collected reflections	749	779	708	750	680
Unique reflections	227	237	225	234	224
h	$-13 \le h \le 13$	$-13 \le h \le 13$	$-13 \le h \le 13$	$-13 \le h \le 13$	$-13 \le h \le 13$
k	$-9 \le k \le 9$	$-9 \le k \le 9$	$-9 \le k \le 9$	$-9 \le k \le 9$	$-9 \le k \le 9$
l	$-9 \le l \le 9$	$-9 \le l \le 9$	$-8 \le l \le 8$	$-9 \le l \le 9$	$-8 \le l \le 8$
$\Delta\rho_{max}$(eÅ^{-3})	2.813	1.113	1.400	1.016	3.907
$\Delta\rho_{min}$(eÅ^{-3})	−1.279	−1.637	−3.367	−3.304	−3.894
Extinction coefficient	0.0031(3)	0.0025(3)	0.0106(6)	0.0052(3)	0.0022(4)
$R(F)_{for}\ F_o^2 > 2\sigma(F_o^2)^a$	0.0338	0.0257	0.0235	0.0235	0.0435
$R_w(F_o^2)^b$	0.0707	0.0619	0.0599	0.0543	0.1137

[a] $R(F) = \Sigma||F_O| - |F_C||/\Sigma|F_O|$.

[b] $R_w(F_0^2)\left[\Sigma\left[w(F_0^2 - F_c^2)^2\right]/\Sigma\left[w(F_0^2)^2\right]\right]^{1/2}$.

Table 3. Atomic parameters, site symmetry and U_{eq} values for $Ln_3Co_4Sn_{13}$ (Ln = Pr, Nd, Sm, Gd, Tb)

Atom	Wyckoff position	x	y	z	U_{eq} (e Å^2)[a]
Pr					
Pr	6*d*	1/4	1/2	0	0.0101(3)
Co	8*e*	3/4	3/4	3/4	0.0076(5)
Sn1	2*a*	0	0	0	0.0154(5)
Sn2	24*k*	0	0.30350(7)	0.15685(7)	0.0135(3)
Nd					
Nd	6*d*	1/4	1/2	0	0.0126(3)
Co	8*e*	3/4	3/4	3/4	0.0089(4)
Sn1	2*a*	0	0	0	0.0169(4)
Sn2	24*k*	0	0.30340(5)	0.15687(5)	0.0148(3)
Sm					
Sm	6*d*	1/4	1/2	0	0.0116(3)
Co	8*e*	3/4	3/4	3/4	0.0088(4)
Sn1	2*a*	0	0	0	0.0209(2)
Sn2	24*k*	0	0.30448(7)	0.15704(7)	0.0141(3)
Gd					
Gd	6*d*	1/4	1/2	0	0.0120(3)
Co	8*e*	3/4	3/4	3/4	0.0082(4)
Sn1	2*a*	0	0	0	0.0120(3)
Sn2	24*k*	0	0.30465(7)	0.15702(6)	0.0138(3)
Td					
Tb	6*d*	1/4	1/2	0	0.0092(5)
Co	8*e*	3/4	3/4	3/4	0.0051(7)
Sn1	2*a*	0	0	0	0.0171(9)
Sn2	24*k*	0	0.30494(12)	0.15689(11)	0.0104(5)

[a] U_{eq} is defined as one-third of the trace of the orthogonalized U_{ij} tensor.

parameters were refined in separate sets of least-squares cycles to determine the correct composition, specifically by checking for defects at the origin on the Sn1 (2*a*) site. The Sn1 sites in $Ln_3Co_4Sn_{13}$ (Ln = Pr-Nd, Sm, Gd, Tb) are fully occupied while their Rh and Ir analogues show partial occupancy on the 2*a* site.[92] Table 3 lists the atomic positions, Wyckoff symmetry, and anisotropic displacement parameters for these compounds.

$Ln_3Co_4Sn_{13}$ (Ln = Pr, Nd, Sm, Gd, Tb) crystallize with the $Yb_3Rh_4Sn_{13}$ structure type.[44] The structure of $Pr_3Co_4Sn_{13}$ is shown in Figure 6 and consists of 3 substructures: Sn icosahedra [Sn1(Sn2)$_{12}$],

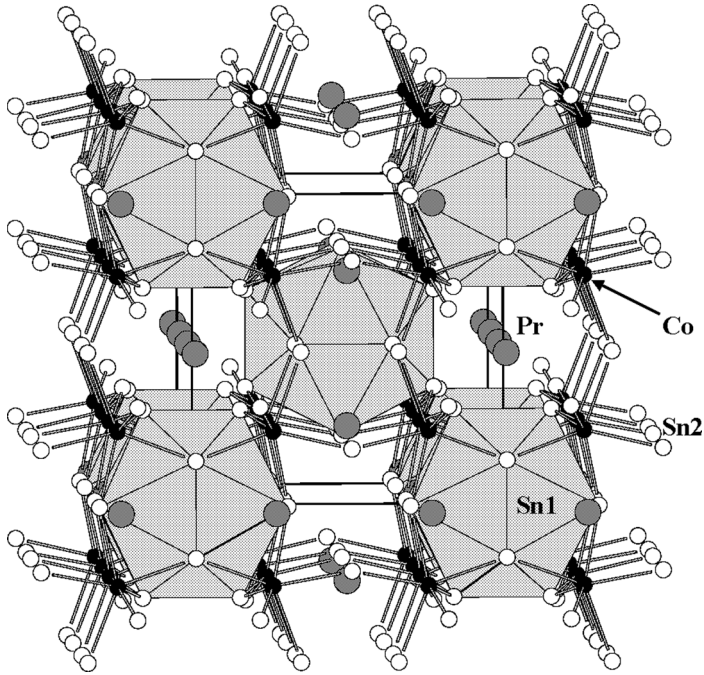

Figure 6. Cubic ($Pm\bar{3}n$) $Pr_3Co_4Sn_{13}$ structure with unit cell and atomic labeling scheme.

Pr cuboctahedra [$Pr(Sn2)_{12}$], and Co trigonal prisms [$Co(Sn2)_6$]. Selected interatomic distances are provided in Table 4. The $Ln_3Co_4Sn_{13}$ structure has been extensively described[90] and compared to the structures of the $A'A''_3B_4O_{12}$-type perovskites. We have observed that in addition to the expected shortening of the inter-atomic distances due to lanthanide contraction, the distances between *Ln*-Sn2 become less distorted within the *Ln*-cuboctahedra as one moves from Pr to Tb, *i.e.*, as the lanthanide atomic radius decreases, the ratio of the two *Ln*-Sn2 distances approaches unity (see Table 4).

Magnetization data are summarized in Table 5. The temperature dependence of the magnetic susceptibility of $Ln_3Co_4Sn_{13}$ (Ln = Pr, Nd, Gd, Tb) in an applied field of 1000 G (0.1 T) is shown in Figure 7. The Sm, Gd, and Tb compounds each order antiferromagnetically below transitions of $T_N \sim 6.6$ K, 12.5 K, and 10.8 K, respectively, while the Pr and Nd compounds display paramagnetic behavior down to the lowest temperature 1.8 K. The effective magnetic moments (μ_{eff}) were obtained by applying a linear fit to the inverse susceptibility data. With the

Table 4. Selected interatomic distances (Å) in $Ln_3Co_4Sn_{13}$(Ln = Pr, Nd, Sm, Gd, Tb)

	Pr	Nd	Sm	Gd	Tb
Sn icosahedron					
Sn1-Sn2 (×12)	3.2735(7)	3.273(3)	3.2666(7)	3.2625(7)	3.2582(2)
Ln cuboctahedron					
Ln-Sn2 (×4)	3.3974(5)	3.398(3)	3.3764(5)	3.3697(4)	3.3061(6)
Ln-Sn2 (×8)	3.3278(7)	3.328(3)	3.3111(6)	3.3061(6)	3.3014(10)
Co trigonal prism					
Co-Sn2 (×6)	2.6073(3)	2.607(2)	2.5957(3)	2.5918(3)	2.5878(4)

exception of Sm, each compound obeyed the Curie-Weiss law ($\chi = C/(T-\theta)$) where C is the Curie constant, T is the temperature and θ is the Weiss temperature. The experimental effective moments are 3.25, 3.51, 7.93, and 10.44 μ_B for the Pr, Nd, Gd, and Tb analogues, respectively. These values are close to the expected values for their Ln^{3+} free ions (see Table 5 under μ_{calc}). This suggests that the Ln^{3+} ions are responsible for the magnetic contributions, and Co does not contribute to the magnetization. It should be noted that the inverse susceptibility of $SnSm_3Co_4Sn_{12}$ does not show linear behavior. This could be attributed to the Van Vleck contribution, in which the contributions of the $J = 7/2$ excited state and the $J = 5/2$ ground state causes the narrowness of the multiplet spacing resulting in a deviation from Curie-Weiss law.[53,93]

Figure 8 shows the field dependence of the magnetization $M(H)$ at 3 K. With increasing field, the Pr, Nd, Sm and Tb compounds each begin

Table 5. Lattice parameters and magnetic data for $Ln_3Co_4Sn_{13}$ compounds[a]

Compound	a (Å)	Ordering, T_N (K)	θ_w (K)	μ_{exp} (μB)	μ_{calc} (μ_B)	Reference
La	9.6430(6)	PM	—	NA	NA	[90]
Ce	9.6022(5)*	PM	−49.5	2.56	2.54	[90]
Pr	9.582(6)	PM	−7.4	3.25	3.58	This work
Nd	9.583(8)	PM	−27.2	3.51	3.62	This work
Sm	9.5350(6)	AFM, 6.6	—	—	0.85	This work
Gd	9.5190(4)	AFM, 12.5	−24.4	7.93	7.94	This work
Tb	9.5010(6)	AFM, 10.8	−17.3	10.44	9.72	This work

[a]Abbreviations: T_N, Néel temperature; θ_w, Weiss temperature; μ_{exp}, experimental effective moment; μ_{calc}, calculated effective moment for Ln^{3+}; PM, paramagnetic; AFM, antiferromagnetic.

*Room temperature data.

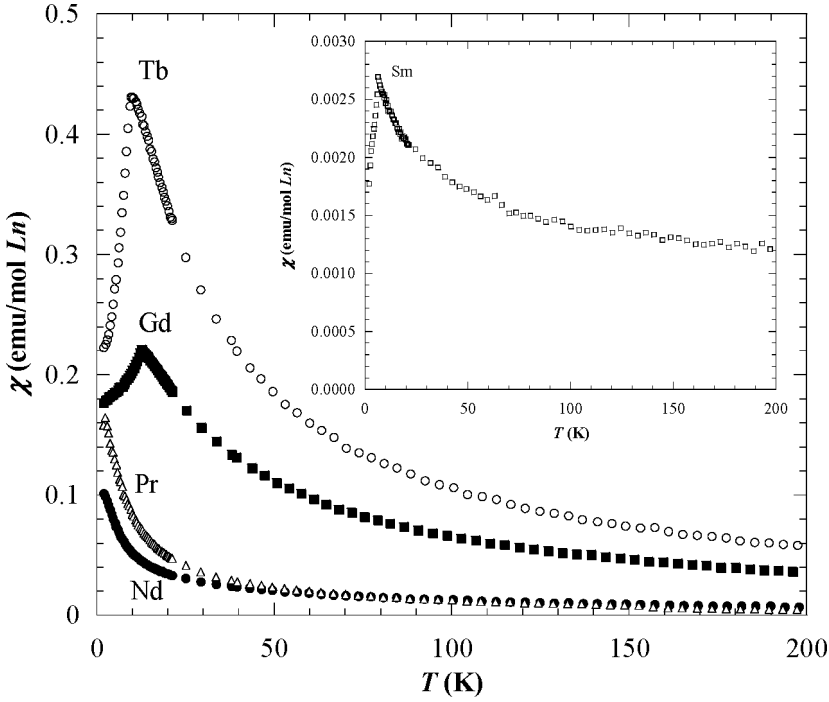

Figure 7. $Ln_3Co_4Sn_{13}$ temperature-dependent susceptibility, $\chi(T)$, in an applied field of $H = 1000$ G for Ln = Pr, Nd, Gd, Tb. Inset: $\chi(T)$ for Sm compound.

to show signs of saturation well below their corresponding saturation moments (μ_s). This is not the case for the Gd compound, whose magnetization increases linearly with increasing field. For reference, the theoretical μ_s values—as calculated from the equation $\mu_s = gJ$, where g is the Landé factor and J is the total angular momentum—for Pr^{3+}, Nd^{3+}, Sm^{3+}, Gd^{3+}, and Tb^{3+} are 3.20, 3.27, 0.71, 7.00, and 9.00 μ_B, respectively. The magnetization behavior observed is also consistent for antiferromagnetic materials. Further investigation of the $Ln_3Co_4Sn_{13}$ structure type will include the examination of other rare earths. In addition, we will further investigate the transport behavior of compounds in this structure type.

SUMMARY AND FUTURE DIRECTIONS

The purpose of this work is to provide sufficient background and motivation for the search for highly correlated materials in order to

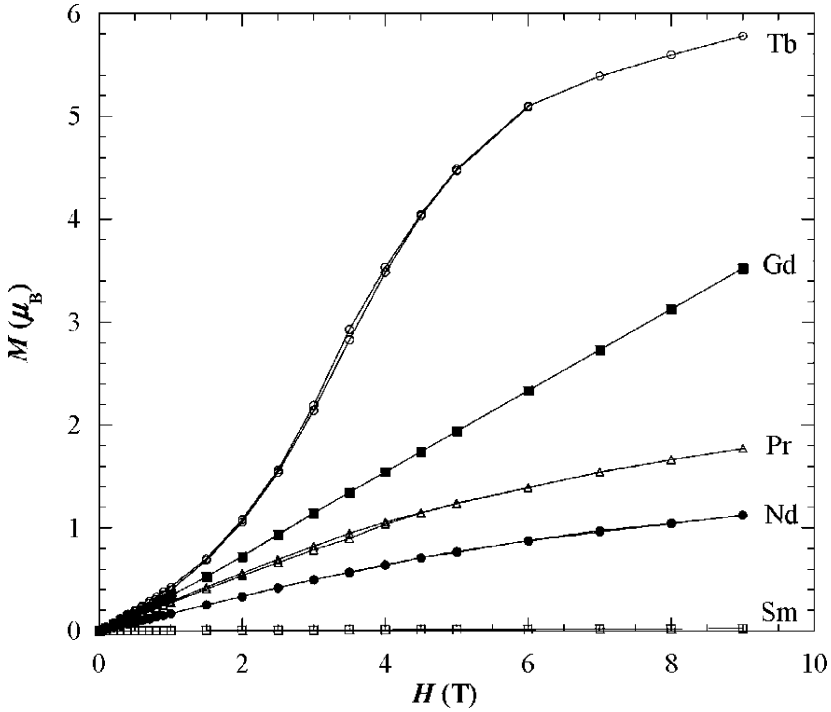

Figure 8. Field-dependent magnetization, $M(H)$, for $Ln_3Co_4Sn_{13}$ (Ln = Pr, Nd, Sm, Gd and Tb).

understand the relationship between crystal chemistry and physical behavior. Our approach, from a chemist's perspective, is driven by studying the structure in terms of dimensionality and local environment of the structural units (*i.e.,* the hybridization from the rare earth atoms and their surrounding neighbors). High quality single crystals are ideal to study the properties of highly correlated systems, and especially vital to understanding the relationship between magnetism and superconductivity, and determining why certain structure types favor heavy-fermion superconductivity.

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