Supporting Information

Materials Screening for the Discovery of New Half-Heuslers: Machine Learning versus Ab Initio Methods

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An additional work on ab initio high throughput

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In addition to the works performed by Carrete *et al.*[1], Ma *et al.*[2], and Zhang *et al.*[3] that are discussed in the main paper, another study from Zhang *et al.*[4] was brought to our attention during the revision stage. In this study, Zhang *et al.* also performed high throughput ab initio calculations to predict new stable compounds, part of them being HHs. Here a comparison between this second study from Zhang *et al.*[4] with those of Carrete *et al.*[1] and Ma *et al.*[2] is provided. The total number of compositions considered for this second study of Zhang *et al.* is 483, 83 of which had already been reported experimentally. The study therefore investigates 400 hypothetical compositions. The chemical compositions of the hypothetical compounds are of type ABC.

5 compositions are simultaneously considered by the three studies. Among them, 4 hypothetical HH compounds are found unstable in all of the three ab initio studies and 1 hypothetical HH compound is found to be stable according to the work of Carrete *et al.* but not in that of Zhang *et al.* and Ma *et al.* (VCoSn).

104 compositions are simultaneously considered by Zhang *et al.* and Carrete *et al.* Out of them, 78 hypothetical HH compounds are found unstable in the two ab initio studies, 12 hypothetical HH compounds are found stable in the two ab initio studies (**ZrNiPb**[5], **ZrPdPb**[4], HfRhBi, **HfIrSb**[4], **ZrRhBi**[4], ZrIrAs, **ZrIrBi**[4], TiIrAs, **TaCoSn**[4], TaRhSn, **TaIrSn**[4], **TaIrGe**[4] – compounds in bold have been successfully synthesized as HHs), 7 hypothetical compounds are found to be stable in the work of Zhang *et al.* but not in that of Carrete *et al.* (*HfNiPb*[5], HfPdPb, HfPtPb, ZrPtPb, ScRhTe, **ZrIrSb**[4], **TiIrSb**[4] – compounds in bold / italicized have been successfully / unsuccessfully synthesized as HHs), and 7 hypothetical HH compounds are found to be stable in the work of Carrete *et al.* but not in that of Zhang *et al.* (ScNiAs, VIrGe, LaRhTe, HfCoBi, AsCoZr, TiCoBi, TiRhBi). According to the work of Zhang *et al.*, the first 2 are stable in another structure than that of the HH and the last 5 are unstable in any structure.

There is no hypothetical HH compound that is simultaneously considered by Zhang *et al.* and Ma *et al.* but that is not considered by Carrete *et al.* The comparison between the works of Ma *et al.* and Carrete *et al.* only is the same as the one given in the main paper.

Performance assessment

The confusion matrix that results from the 10-fold cross-validations is provided in Table S1.

Table S1: Confusion matrix that results from the 10-fold cross-validations.

	Predicted false	Predicted true
Real false	11013	9
Real true	79	85

Predictions

Table S2 lists the chemical compositions for which the machine-learning model gives a probability to be stable in the HH structure higher than 0.5. Table S2 is also available in TableS2.csv.

Table S2: List of the chemical compositions for which the machine-learning model gives a probability to be stable in the HH structure higher than 0.5.

XYZ	Probability	XYZ	Probability	XYZ	Probability	XYZ	Probability
ErNiBi	0.954	MnTcSn	0.717	MgPdTe	0.661	NbCuBi	0.614
TmPtBi	0.947	TaRhSb	0.717	NbPtBi	0.66	EuIrBi	0.614
ErPdBi	0.931	MnZnSb	0.714	TiPtPb	0.659	LiBiTl	0.614
MnRuSb	0.931	TiNiBi	0.713	TaFeBi	0.658	ErAuSb	0.613
TbPtBi	0.913	SmPtBi	0.713	CrCuSb	0.657	TaOsBi	0.613
TbPdBi	0.906	MnPdSn	0.713	MnAuPb	0.656	MgAgBi	0.612
TmPdBi	0.899	MnCoSn	0.709	LiAsAl	0.654	CeCoBi	0.611
EuPdBi	0.89	LuAuPb	0.708	MnPdPb	0.654	MgAuBi	0.609
MnFeSb	0.885	NbNiBi	0.708	YIrBi	0.653	DyCoBi	0.609
LuPtBi	0.882	CrCoSb	0.708	PrPtBi	0.65	TiPdPb	0.609
YPtBi	0.864	CrNiSb	0.708	MnRhPb	0.647	MnNiI	0.608
EuPtBi	0.861	HfPdSb	0.708	MnGaSn	0.646	GdCoBi	0.607
TiRhSb	0.861	ZrCuBi	0.707	MnPtPb	0.645	CrCuTe	0.607
ScPdBi	0.854	TaCoBi	0.706	TmCoBi	0.644	MnNiPb	0.606
MnTeRh	0.846	ScIrBi	0.706	MnPdBi	0.644	MnCuBi	0.604
HfCoBi	0.844	MnInSn	0.706	CrTcSb	0.643	ScRhBi	0.604
LuPdBi	0.831	NbFeBi	0.699	MgNiSn	0.643	TiRhBi	0.604
MnAgSb	0.83	PmPtBi	0.698	ScAuPb	0.642	ZnCuSb	0.604
TiPtSb	0.83	MgCoSb	0.697	TiAuPb	0.642	TaNiBi	0.603
TmAuPb	0.829	ZnCoSb	0.697	ErCoBi	0.642	VMnSb	0.603
VRhSb	0.827	MnFeTe	0.695	MnPtBi	0.642	MgCuTe	0.601
MnRuTe	0.822	TiAuSn	0.692	ScIrSb	0.642	CoVSn	0.6
ZrIrBi	0.819	LuCoBi	0.692	VIrSb	0.642	MnZnTe	0.6
MnTePt	0.819	MgRhSb	0.692	YCuBi	0.641	ScFeBi	0.599
ScPtBi	0.813	NdPtBi	0.688	YbIrBi	0.641	TiCuBi	0.596
PmPdBi	0.812	LiAsIn	0.686	HfSbNi	0.641	MgCoTe	0.595
ZrPdBi	0.803	YbNiBi	0.686	NbRuBi	0.64	ZnCoTe	0.595
MnRhSn	0.803	MnTcTe	0.686	HfPtSb	0.64	MnZnSn	0.594
SmPdBi	0.797	ScRhSb	0.686	MnIrBi	0.638	VRhPb	0.594
MnTcSb	0.797	TiAuSb	0.683	TbIrBi	0.637	TaPtBi	0.593
ZrPtBi	0.795	TiCoBi	0.683	MnNiBi	0.637	ZnNiTe	0.593
ScCoBi	0.786	MnAgTe	0.682	SmNiBi	0.637	MnRhBi	0.591
TbNiBi	0.786	GdIrBi	0.682	MnHgSb	0.636	NbRhBi	0.59
MnNiTe	0.778	ErIrBi	0.681	HoCoBi	0.635	MnIIr	0.59
MnAgSn	0.776	MnCdSb	0.681	MnIPt	0.635	PmLiSn	0.59
ScAuSb	0.776	TiIrBi	0.68	MgGeBi	0.634	MnCoI	0.589
HfIrBi	0.776	ZrRhBi	0.68	VCoTe	0.634	ZnTeFe	0.589
MgAuSb	0.775	HoIrBi	0.679	VAuSb	0.632	MgPtSn	0.589
EuNiBi	0.775	NbPdBi	0.679	LuPdSb	0.632	CrAuPb	0.587
HfNiBi	0.773	VPdSb	0.678	ScAuBi	0.631	PmNiBi	0.587
VCuSb	0.772	TmIrBi	0.677	MnCdSn	0.631	TaIrSb	0.587
MnIrTe	0.772	TiPtBi	0.677	HfIrSb	0.631	ScAgBi	0.586
MnGaSb	0.77	CrFeSb	0.676	NbOsBi	0.628	MnCuSn	0.586
MgPdBi	0.766	MnNiSn	0.676	MnHgSn	0.627	MnVSn	0.586
MnCoTe	0.765	MnCrSn	0.675	CrAgSb	0.626	VAuPb	0.585
CeNiBi	0.761	TiIrSb	0.674	FeVSn	0.626	NbTcBi	0.585
NbCoBi	0.755	LuIrBi	0.673	ZnCrSb	0.625	CrRuSb	0.585
MnCuTe	0.753	MnCrSb	0.671	MgIrSb	0.624	TiRhPb	0.585
TiPdSb	0.753	MgAgSn	0.669	MnRuPb	0.624	MgIrBi	0.583
MnRuSn	0.752	TiCuSb	0.669	MgRuSb	0.623	TiNiTe	0.583
MgCoBi	0.749	CrNiTe	0.668	VNiTe	0.622	ScGeBi	0.582
MnAuTe	0.743	ScCoSb	0.667	MnCoBi	0.621	CrRhSb	0.582
ScCuBi	0.742	MnInSb	0.667	HfFeBi	0.621	MgGaSn	0.582
YCoBi	0.727	TiPdBi	0.666	TiCoTe	0.621	ScRuSb	0.582
MgPtBi	0.727	CrCoTe	0.665	MnIrPb	0.621	HoAuSb	0.581
HfPtBi	0.726	CeIrBi	0.663	MnMoSb	0.618	BeNiSb	0.581
PrPdBi	0.722	HfPdBi	0.663	ZrRuBi	0.616	YbAuPb	0.579
MnFeSn	0.721	DyIrBi	0.662	MnIPd	0.616	BeCuSb	0.579
ZrFeBi	0.719	MgNiTe	0.662	VTcSb	0.615	LiBiHg	0.579
NbIrBi	0.719	TaIrBi	0.661	TmAuSb	0.614	MnOsPb	0.579

XYZ	Probability
MnRuBi	0.578
MgCoSn	0.578
MnPbSn	0.578
NdNiBi	0.577
TaRuBi	0.576
MnSnTe	0.576
MgBBi	0.575
MnAuI	0.574
MnCoPb	0.574
CrPdTe	0.574
MnIRh	0.574
LiBiAu	0.573
LuAuSb	0.573
MgAuPb	0.573
TiRuBi	0.573
VPtSb	0.573
MnAgPb	0.572
LiGeCd	0.572
CrMoSb	0.572
MnGaTe	0.572
TiFeBi	0.571
MnCuI	0.571
MgFeSb	0.57
MnITe	0.569
VPdPb	0.569
MnCuPb	0.567
BeAuSb	0.566
YFeBi	0.565
MnCdTe	0.565
BePtSb	0.564
MnFeI	0.564
MnTeHg	0.564
MnAuBi	0.563
MnPbHg	0.563
MnOsSb	0.563
MnSnSb	0.563
NbRhPb	0.563
VRuPb	0.562
LiGaIn	0.561
VPtPb	0.561
ErAuBi	0.56
TmAuBi	0.56
TaPdBi	0.56
MgPtTe	0.56
BeAuPb	0.559
VNiBi	0.559
VCoBi	0.558
LuFeBi	0.558
MnMoTe	0.558
LiBiCd	0.557
MnFeBi	0.557
CrFeTe	0.557
CrGaSb	0.557
MgRhTe	0.557
MgAgTe	0.556
PrNiBi	0.556
LiSeCd	0.556
EuNiSb	0.556
VIrPb	0.556
BeNiBi	0.555

XYZ	Probability
BePtBi	0.555
MnOsBi	0.555
NbIrPb	0.555
MgGaSb	0.554
NbPtPb	0.554
BeCuBi	0.553
ZrPtSb	0.553
VZnSb	0.553
CrVSn	0.552
MgAsBi	0.551
ZrOsBi	0.551
MnMoSn	0.551
ScRuBi	0.55
CrPdPb	0.55
ZnCrSn	0.55
HoAuBi	0.549
MgGeSb	0.549
LiPbTl	0.549
MnTcPb	0.549
ScAsBi	0.548
VAgSb	0.547
MgZnBi	0.547
MnCrTe	0.547
MgZnSb	0.547
LiBiPb	0.546
CrPtPb	0.546
ZrAuPb	0.545
MnSnI	0.545
AlNiSb	0.544
SmIrBi	0.544
ScCuSb	0.544
MnRuI	0.544
VNiSn	0.544
CrIrPb	0.542
CrRuPb	0.542
HfAuPb	0.541
MnBiHg	0.541
MnInHg	0.541
HfAuSn	0.54
BePdBi	0.54
MnTeSb	0.54
LiBiIn	0.539
BeAuBi	0.538
LuAuBi	0.538
EuCoBi	0.538
CrZnTe	0.538
ErFeBi	0.537
MgIrTe	0.537
MgGeSn	0.536
TiAuBi	0.535
BePdSb	0.534
HoFeBi	0.534
TmFeBi	0.534
HfOsBi	0.534
HfRhBi	0.534
TiIrPb	0.534
NbPdSb	0.534
TbCoBi	0.533
NbPdPb	0.533
VNiPb	0.533

XYZ	Probability
ZrAuBi	0.532
EuAuPb	0.532
BePdTe	0.532
ScZnBi	0.532
MnRhHg	0.532
MnBiAg	0.531
LiGeTl	0.531
BeNiTe	0.53
MgFeBi	0.53
NdIrBi	0.53
VPdBi	0.53
CrRhPb	0.53
MnAgPt	0.529
LiGaAl	0.529
BeCuGe	0.529
MgGaBi	0.529
PmIrBi	0.529
MgRhBi	0.529
TiOsBi	0.529
CrRhTe	0.529
MgHgBi	0.528
CrSbSn	0.528
TiNiPb	0.528
AlCoSb	0.527
VOsSb	0.527
BeCuAs	0.526
CrPdSb	0.526
MnInTe	0.526
MnMoPb	0.526
TiPdSn	0.526
BeAgSb	0.525
YAuBi	0.525
MnTcBi	0.525
VCrSb	0.525
MgHgSb	0.525
MnSbI	0.525
VFeTe	0.524
YbCoBi	0.523
NbReBi	0.523
MnBiMo	0.522
EuPtSb	0.522
TiRhSn	0.522
VIrBi	0.521
VCoPb	0.521
CrNiI	0.521
CrNiPb	0.521
MnFePb	0.521
TaIrPb	0.521
NbAuBi	0.52
MgSiBi	0.52
MnAgHg	0.519
BePtTe	0.519
LaPdBi	0.519
CrCoI	0.519
CrTcTe	0.519
MnIPb	0.519
MnPbTe	0.519
TaPtPb	0.519
CrAgTe	0.518
NbAuPb	0.518

XYZ	Probability
CrMoSn	0.518
CrRuTe	0.517
CeFeBi	0.516
VFeBi	0.516
MnRePb	0.516
MnWPb	0.516
TaRhBi	0.515
CrMoTe	0.515
LiPbIn	0.515
MnPbSb	0.515
BeNiAs	0.514
MnGeSb	0.514
LiPAl	0.513
BeCuTe	0.513
ZrGeBi	0.513
VRhSn	0.513
MnAuHg	0.512
ScHgBi	0.512
NbTcSb	0.512
LiTeAu	0.511
ZnCoI	0.511
TaFeSb	0.511
NbPtSb	0.511
FeIAu	0.51
TiFeTe	0.51
ZnNiI	0.51
MnInPt	0.51
MgPdSn	0.51
MnSnOs	0.51
DyFeBi	0.509
MnPtHg	0.509
NbAuSb	0.508
BeSePd	0.508
ZrRhSb	0.508
NbAgBi	0.507
ZrAgBi	0.507
AlMnSb	0.507
ZrZnBi	0.507
CrRuSn	0.507
MgRhSn	0.507
MnAuRh	0.506
MnCoHg	0.506
HfPtPb	0.506
LiBiBe	0.505
VCuBi	0.505
MnReBi	0.505
ZnCoPb	0.505
TiRhTe	0.505
CrOsPb	0.504
MnGaPb	0.504
NbMnSb	0.504
TiOsSb	0.504
VRuTe	0.504
BePbHg	0.503
LiGeSi	0.503
MnTeOs	0.503
NbNiSb	0.502
NbSnPd	0.502
TiPdTe	0.502
FePdI	0.501
MnRuHg	0.501

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