

Towards Routine GIPAW Calculations for Metallic Systems: the Case of $\text{Sc}T_2\text{Al}$ ($T = \text{Ni},$ $\text{Pd}, \text{Pt}, \text{Cu}, \text{Ag}, \text{Au}$) *Heusler* Phases

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Supporting information

Table S1: Contributions to the isotropic ^{27}Al orbital chemical shieldings (in ppm) and macroscopic susceptibilities (in $10^{-6}\text{cm}^3\text{mol}^{-1}$) from the last five points of the convergence test shown in Fig. 1. The PAW dataset PSL-nl was used with $E_{\text{cut}} = 60$ Ry, $k_bT = 8$ mRy, and $m_s = 0.003 \mu_B$. The values of σ_{macro} were computed from $\chi_{\text{bare}}^{\text{mol}}$ (see text) and the quantities $\bar{\sigma}_s = -1619.93$ ppm, $\mathbf{B}_{\text{ext}} 95.88$ T, and $\mathbf{B}_{F_c} 0.16$ T are already converged.

k-points	σ_o	$\chi_{\text{bare}}^{\text{mol}}$	χ_o^{mol}	σ_{macro}
24984	482.80	-4.54	-3.08	3.80
30420	481.46	-1.78	-0.55	1.46
37926	483.04	-3.66	-2.21	3.06
43560	483.76	-3.99	-2.53	3.33
54720	482.89	-2.76	-1.40	2.30

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Table S2: Contributions to the isotropic ^{27}Al orbital chemical shieldings (in ppm) and macroscopic susceptibilities (in $10^{-6}\text{cm}^3\text{mol}^{-1}$) computed for ScPd_2Al with different k-points samplings in the irreducible BZ. Here, an induced spin magnetic moment $m_s = 1.8 \times 10^{-3} \mu_B$ was used to converge the GS wave functions. The PAW dataset combination PSL-n-n was used with $E_{cut} = 60$ Ry and $k_b T = 8.0$ mRy. The quantities $\bar{\sigma}_s = -595.52$ ppm, $\mathbf{B}_{ext} 12.96$ T, and $\bar{\mathbf{B}}_{Fc} 7.72$ mT are already converged.

k-points	σ_o	χ_{bare}^{mol}	χ_o^{mol}	σ_{macro}
3900	439.48	-49.19	-49.52	10.80
4200	439.54	-46.69	-49.51	10.25
4851	439.62	-48.12	-51.22	10.56
6768	439.37	-49.86	-51.37	10.94

Table S3: Intercepts (a), slopes (b) and the respective standard deviations (Δ) for the dependences of the Fermi contact contributions to the induced magnetic hyperfine field ($\mathbf{B}_{Fc}$) for the six ScT_2Al *Heusler* compounds on the external magnetic field ($\mathbf{B}_{ext}$).

T	a	Δa	b	Δb
Ni	0.079	0.609	0.234	0.026
Pd	0.352	0.567	0.020	0.002
Pt	-0.329	0.988	0.415	0.025
Cu	-0.343	0.095	0.054	0.003
Ag	-0.064	0.103	0.023	0.001
Au	0.059	0.461	0.375	0.022

Table S4: Isotropic ^{27}Al chemical shifts related properties computed for ScT_2Al ($T = \text{Ni}, \text{Pd}, \text{Pt}$) *Heusler* compounds. All the chemical shieldings and shifts are in ppm, the Fermi contact contributions to the induced magnetic hyperfine field ($\mathbf{B}_{Fc}$) are in mT and the external magnetic field ($\mathbf{B}_{ext}$) values are in T.

T	PAW set	$\bar{\sigma}_s$	σ_s	σ_o	$\mathbf{B}_{ext}$	$\bar{\mathbf{B}}_{Fc}$	$\mathbf{B}_{Fc}$	δ_{th}^{comp}	δ_{th}^{comp}
Ni	PSL-n-n	-612.02	-677.13	395.77	12.09	7.40	8.19	778.50	843.61
	PSL-n-sp _n	-612.02	-677.13	395.77	12.09	7.40	8.19	778.50	843.61
	PSL-nl-n	-654.42	-724.54	396.11	11.23	7.35	8.14	818.54	888.66
	PSL-nl-sp _n	-654.42	-724.54	396.11	11.23	7.35	8.14	818.54	888.66
	PSLR-n-n	-612.02	-677.13	421.38	12.09	7.40	8.19	775.04	840.15
	PSLR-n-sp _n	-659.10	-733.89	440.35	11.23	7.40	8.24	803.15	877.94
	JHT	-599.00	-625.04	257.50	12.09	7.24	7.56	746.62	772.66
Pd	PSL-n-n	-581.63	-642.40	439.37	12.09	7.03	7.77	704.51	765.28
	PSL-n-sp _n	-551.25	-607.68	443.59	12.09	6.67	7.35	669.91	726.34
	PSL-nl-n	-538.81	-599.57	439.24	12.96	6.98	7.77	659.80	720.56
	PSL-nl-sp _n	-593.65	-659.10	443.62	11.23	6.67	7.40	710.26	775.71
	PSLR-n-n	-581.63	-646.74	457.85	12.09	7.03	7.82	708.18	773.29
	PSLR-n-sp _n	-594.66	-664.10	462.22	12.09	7.19	8.03	716.84	786.28
	JHT	-577.29	-599.00	303.69	12.09	6.98	7.24	678.72	700.43
Pt	PSL-n-n	-928.88	-1024.37	519.00	12.09	11.23	12.39	972.13	1067.62
	PSL-n-sp _n	-1000.33	-1103.17	532.88	11.23	11.23	12.39	1029.70	1132.54
	PSL-n-sp _{fn}	-1009.68	-1112.52	532.67	11.23	11.34	12.49	1039.26	1142.10
	PSL-nl-n	-1000.33	-1107.84	518.31	11.23	11.23	12.44	1042.25	1149.76
	PSL-nl-sp _n	-1009.68	-1112.52	533.36	11.23	11.34	12.49	1036.55	1139.39
	PSL-nl-sp _{fn}	-937.56	-1037.39	533.05	12.09	11.34	12.55	964.74	1064.57
	PSLR-n-n	-928.88	-1028.71	542.38	12.09	11.23	12.44	970.90	1070.73
	PSLR-n-sp _n	-1009.68	-1121.87	551.94	11.23	11.34	12.60	1042.14	1154.33
	JHT	-928.88	-963.60	369.73	12.09	11.23	11.65	964.27	998.99

Table S5: Isotropic ^{27}Al chemical shifts related properties computed for $\text{Sc}T_2\text{Al}$ ($T = \text{Cu}, \text{Ag}, \text{Au}$) *Heusler* compounds. All the chemical shieldings and shifts are in ppm, the Fermi contact contributions to the induced magnetic hyperfine field ($\mathbf{B}_{Fc}$) are in mT and the external magnetic field ($\mathbf{B}_{ext}$) values are in T.

T	PAW set	$\bar{\sigma}_s$	σ_s	σ_o	$\mathbf{B}_{ext}$	$\bar{\mathbf{B}}_{Fc}$	$\mathbf{B}_{Fc}$	δ_{th}^{comp}	δ_{th}^{comp}
Cu	PSL-n-dn	-93.49	-102.84	277.45	11.23	1.05	1.15	378.29	387.64
	PSL-n-spfn	-82.47	-95.49	271.20	12.09	1.00	1.15	373.52	386.54
	PSL-nl-dn	-91.15	-104.17	277.18	12.09	1.10	1.26	374.20	387.22
	PSL-nl-spfn	-98.16	-112.19	272.46	11.23	1.10	1.26	385.93	399.96
	PSLR-n-dn	-95.49	-108.51	293.17	12.09	1.15	1.31	386.72	399.74
	PSLR-n-spfn	-91.15	-104.17	291.32	12.09	1.10	1.26	384.23	397.25
	JHT	-91.15	-95.49	122.81	12.09	1.10	1.15	373.46	377.80
Ag	PSL-n-n	-69.45	-78.13	302.85	12.09	0.84	0.94	328.85	337.53
	PSL-n-spfn	-91.15	-104.17	298.16	12.09	1.10	1.26	355.24	368.26
	PSL-nl-n	-104.17	-121.54	302.65	12.09	1.26	1.47	361.75	379.12
	PSL-nl-spfn	-108.51	-121.54	297.99	12.09	1.31	1.47	370.75	383.78
	PSLR-n-dn	-104.17	-121.54	321.91	12.09	1.26	1.47	366.66	384.03
	PSLR-n-spfn	-108.51	-121.54	320.58	12.09	1.31	1.47	372.33	385.36
	JHT	-91.15	-95.49	147.76	12.09	1.10	1.15	348.51	352.85
Au	PSL-n-n	-476.79	-528.21	400.18	11.23	5.35	5.93	638.86	690.28
	PSL-n-spfn	-486.14	-537.56	395.40	11.23	5.46	6.04	652.99	704.41
	PSL-n-spfn	-495.49	-546.91	389.06	11.23	5.56	6.14	668.68	720.10
	PSL-nl-n	-481.47	-532.89	400.22	11.23	5.41	5.98	641.48	692.90
	PSL-nl-spfn	-490.82	-542.24	395.51	11.23	5.51	6.09	655.54	706.96
	PSL-nl-spfn	-500.17	-551.58	389.18	11.23	5.62	6.19	671.22	722.63
	PSLR-n-dn	-481.47	-537.56	419.69	11.23	5.41	6.04	646.18	702.27
	PSLR-n-spfn	-486.14	-542.24	398.01	11.23	5.46	6.09	672.53	728.63