Modified Becke-Johnson potential investigation of half-metallic Heusler compounds

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We have investigated the electronic structures of various potentially half-metallic Heusler compounds with the Tran-Blaha modified Becke-Johnson + local density approximation (TB-mBJLDA) potential within the density functional theory. The half-metallic gaps are considerably enhanced with respect to values from the Perdew-Burke-Ernzerhof (PBE) functional. In particular the unoccupied densities of states are modified by the mBJLDA potential, and agreement with experiment is considerably worse than for PBE results. The agreement of the densities of states can be improved by reducing the Tran-Blaha parameter c. However, ground-state properties such as the hyperfine fields are more accurately described by the PBE functional than by the mBJLDA. Despite its success for ionic and covalent semiconductors and insulators, we conclude that the mBJLDA is not a suitable approximation for half-metallic Heusler compounds.

I. INTRODUCTION

The half-metallic Heusler compounds[1–4] have frequently been considered as ideal electrode materials for spintronic devices. High tunnel and giant magnetoresistance and high spin injection efficiency are expected from half-metals, i.e., materials that have full spin polarization at the Fermi energy. Heusler compounds are ternary intermetallic compounds with the general chemical formula X_2YZ, where X and Y are transition metals, and Z is a main group element. They form the cubic $L2_1$ structure, which has inversion symmetry and belongs to space group $Fm\bar{3}m$. Some of the closely related inverse Heusler compounds with the Hg_2CuTi prototype structure (space group $F\bar{4}3m$) have only recently been discovered to exhibit half-metallic ferromagnetism as well.[5,6] The half-metals from both classes of compounds follow the Slater-Pauling rule, which relates the magnetic moment m [given in μ_B per formula unit (f.u.)] and the number of valence electrons N_V via $m = N_V − 24$.[3]

Most electronic structure studies of Heusler compounds are based on the Kohn-Sham framework of density functional theory[7,8] (DFT), which is today the main tool to obtain the electronic structure of solids. However, a well-known failure of this framework is the underestimation of band gaps. This is closely related to a missing derivative discontinuity Δ_{xc} in the approximate exchange-correlation (xc) functionals. However, the Kohn-Sham gap ϵ_g differs from the true gap E_g by this discontinuity even for the exact xc functional, which has to be computed and added to the Kohn-Sham gap "by hand."[9] This is in principle also true for the gap of half-metals.[10]

The appropriate framework to discuss band gaps is the many-body perturbation theory, e.g., within the GW approximation.[11] Unfortunately, this approach is computationally very expensive. Tran and Blaha recently proposed an alternative, equally accurate, and computationally cheaper method to obtain the gap directly as differences of Kohn-Sham eigenvalues: they modified the Becke-Johnson exchange potential[12] with an additional parameter c, so that it reads[13]

$$v_{x,\sigma}^{mBJ}(\mathbf{r}) = cv_{x,\sigma}^{BR}(\mathbf{r}) + (3c - 2)\frac{1}{\pi}\sqrt{\frac{5}{12}}\sqrt{\frac{2t_\sigma(\mathbf{r})}{n_\sigma(\mathbf{r})}}, \qquad (1)$$

where $n_\sigma(\mathbf{r})$ is the spin-dependent electron density and $t_\sigma(\mathbf{r})$ is the spin-dependent kinetic-energy density. $v_{x,\sigma}^{BR}(\mathbf{r})$ is the Becke-Roussel potential, which models the Coulomb potential created by the exchange hole.[14] Due to the kinetic-energy-dependent term in the modified Becke-Johnson (mBJ) potential, it reproduces the step structure and derivative discontinuity of the effective exact exchange potential of free atoms.[15] The parameter c was proposed to be determined self-consistently from the density by

$$c = \alpha + \beta \left(\frac{1}{V_{cell}} \int_{cell} \frac{|\boldsymbol{\nabla}n(\mathbf{r}')|}{n(\mathbf{r}')} d^3r' \right)^{1/2}, \qquad (2)$$

with two parameters α,β, which have been chosen to fit the band gaps of a broad range of solids. It can be related to the dielectric response of the system.[16,17] c increases with the gap size and has a typical range of 1.1–1.7.[13] The mBJ potential has been proposed to be combined with local density approximation (LDA) correlation (mBJLDA). Its particular merits and limits have been reviewed by Koller *et al.*[18] In a recent paper, Koller *et al.* have suggested a new and more balanced parametrization of c, based on a larger test set of solids.[19] This reparametrization gives, however, rather similar results as the original parametrization for small-gap materials. Making use of the kinetic-energy density, the mBJ potential is formally a meta generalized gradient approximation (GGA) potential.[20]

In a recent paper, Guo and Liu have used the mBJLDA potential to investigate the half-metallic ferromagnetism of zinc-blende transition-metal pnictides and chalcogenides. They found that the mBJLDA enhances the half-metallic gaps significantly with respect to conventional DFT calculations.[21] In the present paper, we aim to investigate if the half-metallic gap of Heusler compounds is enhanced with the mBJLDA, and if such an enhancement leads to an improved description of the electronic structure.

II. COMPUTATIONAL DETAILS

All calculations in this work are based on the full-potential linearized augmented-plane-wave (FLAPW) method. The mBJLDA calculations are done with the ELK code.[22] The

mBJ exchange potential is available through an interface to the LIBXC library.[23] Γ-centered $21 \times 21 \times 21$ **k**-point meshes are used with 286 points in the irreducible wedge for Heusler compounds and 506 points for the inverse Heusler compounds. A Gaussian smearing of 1 mHa is applied in all calculations. The muffin-tin radii are 2.0 bohr, and the momentum cutoff for the plane-wave expansion is $k_{max} = 4.0$ bohr^{-1}. The angular momentum expansion of potential and wave functions is taken to $l_{max} = 10$. The mBJ exchange potential is coupled with the Perdew-Wang LDA correlation.[24] For comparison with results from a generalized gradient approximation, we choose the Perdew-Burke-Ernzerhof (PBE) functional,[25] which is the standard functional in most current studies of Heusler compounds. All calculations are based on experimental lattice constants.

III. RESULTS

A. Gaps and magnetic moments

We have chosen the Heusler compounds for our study along the lines of Ref. 4, and we added some (inverse) Heusler compounds of current interest.[5,6,26] The main results of our calculations are collected in Table I, which displays the mag-netic moments and band gaps within the PBE and mBJLDA calculations, and the Tran-Blaha parameters c. Notably, the c parameter is always in the range 1.09–1.20. The corresponding density of states (DOS) plots are shown in Fig. 1.

We observe that all materials that are half-metallic with the PBE functional are half-metallic in the mBJLDA as well, with an increased gap. Some cases which have a gap in PBE calculations with the Fermi energy located outside the gap (marked by asterisks) become half-metals in the mBJLDA (Co$_2$VAl, Co$_2$CrGa, Co$_2$FeAl, Co$_2$FeGa, and Mn$_2$VGa). Co$_2$MnAl, Co$_2$MnSn, Co$_2$FeSi, Co$_2$FeGe, and Ru$_2$MnSb have a larger gap in the mBJLDA, but the Fermi energy remains outside the gap.

Mn$_2$VAl and Mn$_2$VGa are ferrimagnetic Heusler compounds. Mn$_2$VAl is characterized as a half-metal with a majority gap by PBE calculations, and the mBJLDA has only weak influence on the size of this gap. This goes along with the smallest value of c among the materials studied here. Mn$_2$VGa has a pseudogap around the Fermi level with the PBE functional. A gap is opened in the mBJLDA, and the Fermi energy is located within the gap. Fe$_2$VAl, Fe$_2$VGa, and Fe$_2$TiSn, three semimetals or zero-gap semiconductors within the PBE approximation, are predicted to be semiconductors

TABLE I. Magnetic moments and (half-metallic) band gaps computed with PBE and mBJLDA (marked as mBJ) methods at the given experimental lattice constants. Lattice constants and experimental magnetic moments are taken from References 2,4–6,26–31. Moments are given in μ_B/f.u., gaps are given in eV, lattice constants are given in Å. The parameter c is dimensionless. Asterisks mark gaps which are above or below the Fermi energy.

|	N_V	a_{expt}	m_{expt}	m^{PBE}	E_g^{PBE}	m^{mBJ}	E_g^{mBJ}	c
			Co$_2$YZ Heusler compounds					
Co$_2$TiAl	25	5.85	0.74	1.00	0.40	1.00	1.11	1.12
Co$_2$TiSn	26	6.08	1.96	2.00	0.47	2.00	1.16	1.17
Co$_2$VAl	26	5.72	1.95	2.00	0.36*	2.00	0.65	1.13
Co$_2$ZrSn	26	6.25	1.81	2.00	0.50	2.00	1.50	1.16
Co$_2$CrGa	27	5.81	3.01	3.04	0.39*	3.00	1.06	1.18
Co$_2$MnAl	28	5.75	4.04	4.03	0.61*	4.04	1.29*	1.14
Co$_2$MnSi	29	5.65	4.97	5.00	0.81	5.00	1.42	1.15
Co$_2$MnGe	29	5.75	4.93	5.00	0.57	5.00	1.49	1.19
Co$_2$MnSn	29	5.98	5.08	5.03	0.39*	5.04	1.36*	1.19
Co$_2$FeAl	29	5.73	4.96	4.99	0.06*	5.00	0.75	1.14
Co$_2$FeGa	29	5.74	5.04	5.02	0.02*	5.00	0.80	1.20
Co$_2$FeSi	30	5.64	5.97	5.47	0.11*	5.79	0.82*	1.16
Co$_2$FeGe	30	5.74	5.90	5.63	0.09*	5.98	0.90*	1.20
			Other Heusler compounds					
Mn$_2$VAl	22	5.92	1.94	2.00	0.28	2.00	0.48	1.09
Mn$_2$VGa	22	5.91	1.88	1.99	0.02*	2.00	0.27	1.15
Fe$_2$VAl	24	5.76	0.00	–	–	–	0.31	1.12
Fe$_2$VGa	24	5.78	0.00	–	–	–	0.39	1.17
Fe$_2$TiSn	24	6.09	0.00	–	–	–	0.69	1.16
Ru$_2$MnSb	28	6.20	4.40	4.03	0.28*	4.06	0.44*	1.19
Ni$_2$MnSn	31	6.05	4.05	4.03	–	4.17	–	1.19
Cu$_2$MnAl	32	5.95	3.60	3.51	–	3.50	–	1.13
Cu$_2$MnSn	33	6.17	4.11	3.86	–	3.91	–	1.19
			Inverse Heusler compounds					
Cr$_2$CoGa	24	5.80	0.35	0.08	0.19*	0.03	0.66*	1.17
Mn$_2$CoAl	26	5.84	1.95	2.00	0.43	2.00	0.68	1.12
Mn$_2$CoGe	27	5.80	2.99	3.00	0.36	3.00	0.76	1.18
Fe$_2$CoSi	29	5.65	4.99	4.96	–	5.00	0.57	1.16
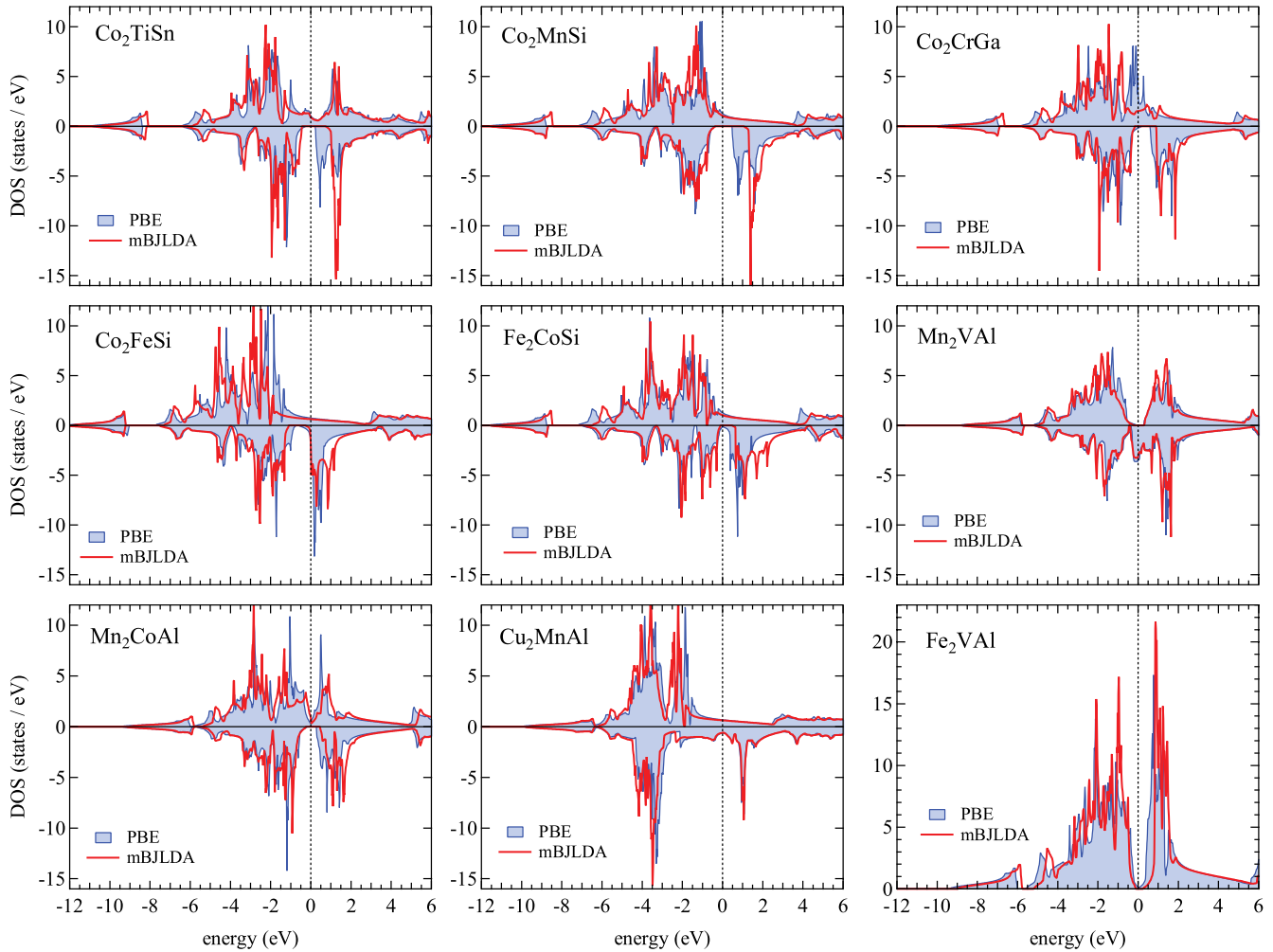
FIG. 1. (Color online) Density of states plots for selected Heusler and inverse Heusler compounds. Shaded blue areas correspond to PBE calculations; solid red lines represent mBJLDA calculations. Spin majority states are positive, minority states are negative. The energy scale is given with respect to the Fermi energy.

by the mBJLDA. Do *et al.* investigated Fe_2VAl with hybrid functionals and $DFT + U$ and also found a semiconductor with an energy gap depending on the choice of parameters.[32] They also report an mBJLDA calculation with a gap of 0.22 eV, which is somewhat smaller than our value.

Ni_2MnSn, Cu_2MnAl, and Cu_2MnSn were included in this study to observe the influence of the mBJLDA on the magnetic moment and the exchange splitting of ferromagnetic Heusler compounds that do not have a gap at all. In all three cases the magnetic moments obtained with the PBE functional and the mBJLDA are close; while the agreement with experiment is very good for Ni_2MnSn, both approximations underestimate the moment of Cu_2MnAl and Cu_2MnSn. This is surprising, seeing that the mBJLDA predicts too large moments for Fe ($2.49\mu_B$) and Ni ($0.74\mu_B$) and too large exchange splittings at the same time.[18] Notably, the PBE functional (and the LDA) already predict too large exchange splittings, although the moments are accurate.[33] The small influence of the mBJLDA on the magnetic moment in Cu_2MnAl and Cu_2MnSn may be associated with the localized character of the magnetic moment.[2] Thus, an increase of the exchange splitting does not

lead to an enhanced magnetic moment. In contrast, Fe and Ni have more itinerant character, so the moments depend strongly on the magnitude of the exchange splitting.

The inverse Heusler compound Cr_2CoGa has been predicted as a nearly fully compensated ferrimagnet.[34] It has a small gap slightly above the Fermi energy. In the mBJLDA description, this gap is further enlarged and the Fermi energy moves closer to the gap, thereby further reducing the magnetic moment towards zero. Experimentally, a nonzero moment is observed,[26] which may be due to atomic disorder. Mn_2CoAl is a ferrimagnet, and it is predicted to be a spin-gapless semiconductor[35] by use of the PBE functional. This prediction has recently been confirmed experimentally.[36] This is also the case within the mBJLDA, and the minority spin gap is only slightly enhanced (similarly to the case of Mn_2VAl). Mn_2CoGe is a half-metal in both approximations, but the mBJLDA considerably enhances the minority gap. Fe_2CoSi is described by the PBE approximation as a conventional ferromagnet with a pseudogap in the minority spin channel around the Fermi energy. The mBJLDA opens a sizable gap and predicts Fe_2CoSi to be a half-metal.

We note that the magnetic moments inside the muffin-tin spheres are increased with the mBJLDA in all cases, which is compensated by an antiparallel interstitial moment (for ferromagnets) or by the antiparallel alignment of the muffin-tin moments (for ferrimagnets). The integer magnetic moments are protected by the half-metallic gaps.

B. Densities of states

In Fig. 1 we compare the DOSs from PBE and mBJLDA calculations for some selected compounds. We observe that the effect of the mBJLDA (compared to the PBE approximation) is very material dependent and nontrivial. In all cases we observe that the energy range of the occupied d states is compressed; the d-band minima are raised and the states close to the Fermi energy are lowered. The exchange splittings of the occupied d states are enhanced in all cases. The low-lying s states from the sp element are shifted up in some cases, or remain at the PBE position. We further note that the effects on spin majority and minority states are quite different. The occupied minority states are least affected in most cases, while the occupied majority states and the unoccupied minority states show somewhat larger changes.

The enhancement of the half-metallic gaps is visible for all compounds. Mn_2VAl shows only slight changes with the mBJLDA compared to the PBE calculation, which is partly due to the low value of c. In the case of Co_2FeSi we see that the Fermi energy is located slightly above the bottom of the minority conduction-band minimum in both cases.

C. Dependence on c

To estimate the influence of the mBJ model parameter c, we calculated the gaps of Co_2TiSn, Co_2MnSi, Co_2FeSi, Mn_2VAl, Fe_2VAl, and Mn_2CoGe with different fixed values of c. The results are displayed in Fig. 2 (top). We can identify two classes of dependencies on c within the range of our investigation: an approximately linear one (Fig. 2, top left), and one that saturates at rather low c values (Fig. 2, top right). Remarkably, the three compounds belonging to the second class are ferrimagnets, whereas the compounds in the first class are ferromagnets or nonmagnetic (Fe_2VAl). In contrast to semiconductor and insulator gaps,[13] the half-metallic gap does not necessarily grow monotonically as c is increased.

Setting $c \approx 0.95$ nearly restores the PBE values for the gaps in most cases. Also the densities of states agree very well with those from the PBE approximation with $c = 0.95$. This agrees with the finding by Koller *et al.* that the magnetic moment of Fe can be tuned to the PBE value with c slightly lower than unity.[18] However, they also mention that the calculated hyperfine fields at the Fe sites are much worse with a small c, which indicates subtle differences in the description of the electronic structures, in particular of the s states. The original Becke-Johnson exchange potential (i.e., $c = 1$) gives band structures very similar to those obtained with the PBE functional, but with a slightly larger gap. A corresponding DOS plot is shown exemplarily for Co_2MnSi in Fig. 2 (bottom). This agrees with earlier calculations with the BJLDA potential of semiconductor and insulator gaps.[37]
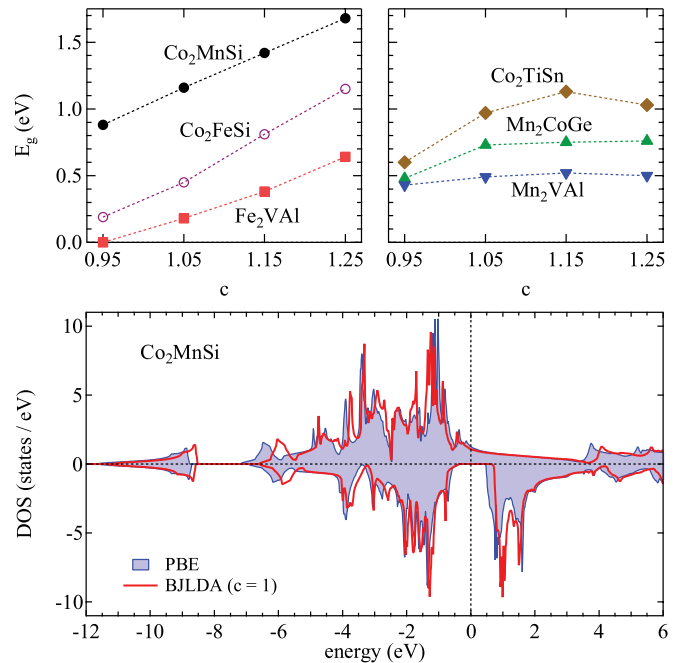

FIG. 2. (Color online) Top: Dependence of the half-metallic and semiconducting gaps on the mBJ model parameter c. Co_2FeSi (open symbols) is not a half-metal for the plotted range of c. Bottom: DOS plots for Co_2MnSi with the PBE approximation and the BJLDA ($c = 1$).

It is well known that the local xc functionals (in both the LDA and the GGA) fail to predict the ground-state magnetic moment of Co_2FeSi correctly.[27] The mBJLDA does not resolve this problem with the c value from Eq. (2). With $c = 1.35$ a just half-metallic ground state can be obtained with the Fermi energy at the minority conduction-band minimum, and the magnetic moment is $6\mu_B$, in accordance with experiments.[27] As we discuss in the following, such a high value of c is rather unrealistic.

D. Comparison to experiments

The experimental determination of the half-metallic gap is notoriously difficult. Only indirect evidence for the presence and the size of such gaps is available. This evidence comes from tunnel spectroscopy of magnetic tunnel junctions with amorphous barriers[38–40] or from x-ray absorption spectroscopy.[41,42] For Co_2MnSi, a combination of the minority spin-flip gap from tunnel spectroscopy of 0.25–0.35 eV and the position of the x-ray absorption maximum of Co at its L_3 edge of 0.9 ± 0.1 eV above E_F gives an upper boundary of 1.2 ± 0.1 eV for the minority gap. Note, however, that the Co d DOS maximum is about 0.3 eV above the minority conduction-band minimum in the PBE and mBJLDA calculations; the gap is therefore smaller than 0.9 ± 0.1 eV. The x-ray absorption spectrum does not directly reflect the unoccupied DOS because of the formation of an exciton (due to the core hole) in the absorption process. The exciton binding energy was estimated to be 0.5 eV in Ref. 41 and is taken into account in the above discussion. Thus, considering the available experimental data, the mBJLDA value for the minority gap is considerably too large and the PBE value is slightly too small.
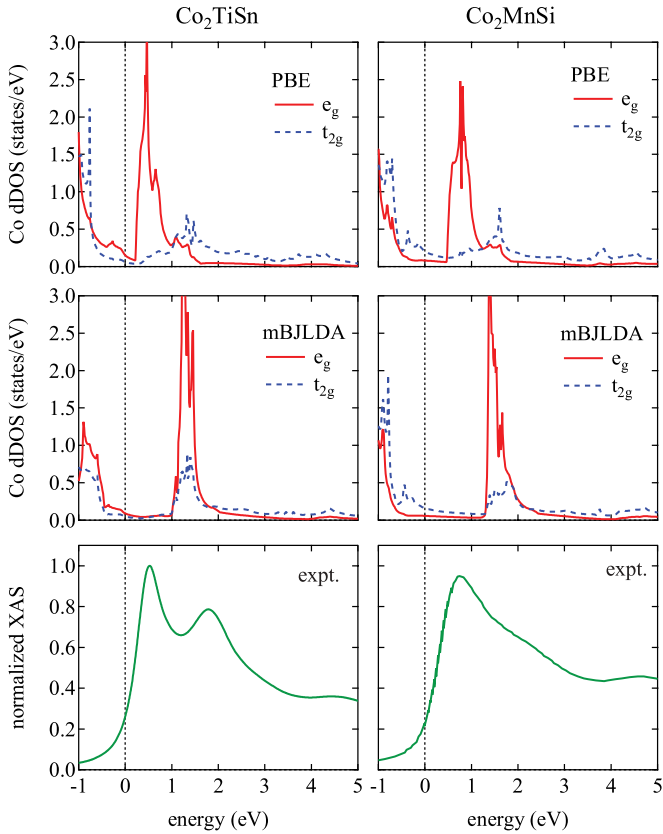
FIG. 3. (Color online) Experimental Co L_3 absorption spectra of Co$_2$TiSn and Co$_2$MnSi (bottom row) and corresponding d DOSs with PBE approximation and mBJLDA. Data taken from Refs. 43 and 44.

TABLE II. Hyperfine contact fields (in kOe) and site-resolved magnetic moments (in μ_B) of chosen Heusler compounds. See text for mean (absolute) errors.

	PBE		mBJLDA		BJLDA		Expt.
	H_{hf}	m	H_{hf}	m	H_{hf}	m	H_{hf}
Co$_2$MnSi							
Co	−108	1.07	−139	1.18	−45	1.12	−145[a]
Mn	−219	2.85	−154	3.01	−51	2.91	−336[a]
Si	26	−0.04	60	−0.06	40	−0.05	–
Co$_2$MnSn							
Co	−139	1.00	−200	1.17	−83	1.06	−156[a]
Mn	−224	3.02	−178	3.19	−44	3.06	−344[a]
Sn	89	−0.03	288	−0.04	133	−0.03	105[a]
Co$_2$TiSn							
Co	52	1.05	14	1.27	106	1.09	21[b]
Ti	−82	−0.03	−137	−0.19	−85	−0.04	–
Sn	94	0.00	331	0.01	155	0.01	82[b]
Mn$_2$VAl							
Mn	−76	1.45	7	1.59	26	1.46	−99[c]
V	−100	−0.84	−314	−1.01	−249	−0.85	−64[c]
Al	−31	−0.02	−37	−0.03	−31	−0.03	−25[c]
Cu$_2$MnSn							
Cu	−234	0.06	−219	0.05	−190	0.04	−175[b]
Mn	−79	3.44	−4	3.62	139	3.41	–
Sn	488	0.00	535	0.00	539	0.00	196[b]

[a] Reference 49.
[b] Reference 28.
[c] Reference 50.

Also, the spectral shapes of the Co L_3 x-ray absorption spectra are not well reproduced by the mBJLDA. Consider, e.g., Co$_2$TiSn (Fig. 3, left), which has a pronounced double-peak structure[43] arising from a pure Co e_g and a Co-Ti hybrid t_{2g} state, which are separated by 0.9 eV in the PBE calculation. Due to the excitation process, a screened core hole is formed and pulls the localized e_g states down by 0.3 eV,[43] so the total separation of the peaks is predicted to be 1.2 eV, which is close to the experimental value of 1.3 eV (see Fig. 3). Note that the experimental spectra are broadened due to finite-lifetime effects. This double-peak structure is clearly visible in the unoccupied PBE DOS of Co$_2$TiSn in Fig. 3. In contrast, the mBJLDA moves the e_g peak up, such that it overlaps with the t_{2g} peak, and no double-peak structure would be visible. A similar but less pronounced structure is also present in the absorption spectra of Co$_2$MnSi (Fig. 3 right),[44] which is reproduced by the PBE calculation but is not present in the mBJLDA calculation for the same reason as for Co$_2$TiSn.

The gaps of Fe$_2$VAl and Fe$_2$TiSn predicted by the mBJLDA should easily be detectable with optical methods and by electrical transport. However, both compounds have been characterized as semimetals by experiments.[45–47] Hence, the overestimation of the Heusler compound band gaps by the mBJLDA is not limited to magnetic cases, where the different spin densities may lead to an error in the determination of c, but is also found for paramagnetic materials.

While the band gap (and actually the entire band structure) is a property of excited states, the magnetic moments and hyperfine fields are ground-state properties. The total moments were already discussed above and good agreement with experiment was shown by PBE and mBJLDA results in most cases. We now turn to the hyperfine fields H_{hf}, which we calculate from the Fermi contact term, ignoring other contributions.[48] As mentioned above, the BJLDA potential ($c = 1$) provides densities of states that are very similar to those from PBE calculations. Therefore we compare the hyperfine contact fields computed with the PBE, mBJLDA, and BJLDA methods for five Heusler compounds. The hyperfine fields and the corresponding site-resolved magnetic moments are given in Table II. The PBE functional provides reasonable hyperfine contact fields with a mean absolute error (mean error) of 64 kOe (+44 kOe) for the present test set. No clear trend towards an over- or underestimation of the hyperfine fields is visible. The PBE values are similar to those calculated by Picozzi *et al.*[49] The mBJLDA and BJLDA perform less well, with mean absolute errors (mean errors) of 133 kOe (+73 kOe) and 135 kOe (+101 kOe), respectively, where the mean of the experimental values is −78 kOe. Since the mBJLDA and BJLDA produce very different results, the hyperfine fields depend critically on the value of c. Again, no clear trend towards an over- or underestimation with respect to experiment or with respect to the other approximations is obvious. Although the BJLDA and PBE calculations predict similar magnetic moments and DOSs, there are notable differences in the wave functions, particularly those of the s states, which provide the dominant contribution to the transferred hyperfine

TABLE III. Total contact hyperfine fields and valence and core contributions for Co_2MnSi from PBE, mBJLDA, and BJLDA calculations. All values given in kOe.

	PBE			mBJLDA			BJLDA		
	Total	Valence	Core	Total	Valence	Core	Total	Valence	Core
Co	−108	32	−140	−139	−61	−78	−45	1	−46
Mn	−219	152	−371	−154	55	−209	−51	79	−130
Si	26	28	−2	6	62	−2	40	41	−1

contact field. The PBE functional performs best in this respect.

To give some more insight into the origin of the hyperfine fields, we break them down into core and valence contributions for Co_2MnSi, where the Co and Mn $3s$ states are treated as core levels (Table III). Here one finds that both the core and valence contributions differ strongly among the three potentials. It has been shown that the core contribution scales in direct proportion to the magnetic moment within the muffin-tin spheres.[49] Here, we find on average −130 kOe/μ_B for PBE, −68 kOe/μ_B for mBJLDA, and −43 kOe/μ_B with BJLDA calculations, with little difference between Co and Mn. However, Novák *et al.* have shown by applying a self-interaction-corrected potential to the core states, that the core contribution should actually be *larger* in magnitude than that obtained with the PBE functional.[51] Thus, the (m)BJLDA values are clearly worse than the PBE values.

The mBJLDA potential was originally designed as an empirical means to obtain the band gap directly as differences of Kohn-Sham eigenvalues.[52] Thus, the overall band structure is not intended to be improved over other semilocal approximations, such as the PBE functional. In other words, the mBJLDA potential is neither designed to be a better approximation to the quasiparticle self-energy, nor to be a better approximation to the true Kohn-Sham potential,[52] which is reflected in the worse description of the hyperfine fields. As shown by Kresse *et al.*, the mBJLDA band dispersions can be even worse than those of the PBE approximation, for example in the case of optical absorption spectra of SiO_2.[53] They also point out that a local potential cannot simultaneously predict the gap and the band dispersions correctly. On the other hand, the mBJLDA seems to be beneficial for the local magnetic moments of strongly correlated materials[18] and for the oxygen K-edge energy-loss near-edge structure (ELNES) description of NiO.[54]

Interestingly, the mBJLDA fails for (metallic) itinerant-electron systems in a similar way as do the hybrid functionals, which mix a fraction of Hartree-Fock exchange with the usual semilocal exchange.[18,55–57] Hybrids predict too large bandwidths (which does not happen with the mBJLDA)

and much too large magnetic moments. In particular, the hybrid PBE0 functional is an unsuitable approximation for the Co_2YZ Heusler compounds, predicting much too large band gaps and exchange splittings.[58] This arises from an imbalance between the exchange and correlation parts of the potential. A remedy for that is to combine the exact exchange with correlation derived from many-body perturbation theory,[9,33,59] but such schemes are not widely used and computationally extremely expensive. However, no correlation potential to properly complement the mBJ exchange is known.

IV. CONCLUSIONS

We have calculated magnetic moments and densities of states of 26 (inverse) Heusler compounds with the Tran-Blaha modified Becke-Johnson + LDA potential. In the half-metallic cases, the gaps are much wider than those obtained with the PBE functional. We have shown for some cases (for which sufficient experimental data are available) that the mBJLDA does not improve the description of the half-metallic band gaps or band structures of these compounds with respect to the PBE functional. The original Becke-Johnson potential + LDA predicts similar band structures as the PBE functional, but with a slightly larger gap. Also the magnetic moments agree well with PBE results. Such a description does seem to be more reasonable for materials with a metallic dielectric response, whereas highly correlated magnetic insulators have been shown to be overall better described by the mBJLDA potential.[18,54] On the other hand, the hyperfine fields (as an important ground-state property) predicted by the mBJLDA and BJLDA are worse than those from the PBE approximation.

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