Quantum-Many-Body Intermetallics: Phase Stability of Fe₃Al and Small-Gap Formation in Fe₂VAl

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Various intermetallic compounds harbor subtle electronic correlation effects. To elucidate this fact for the Fe-Al system, we perform a realistic many-body investigation based on the combination of density functional theory with dynamical mean-field theory in a charge self-consistent manner. A better characterization and understanding of the phase stability of bcc-based D₀₂₋ Fe₃Al through an improved description of the correlated charge density and the magnetic-energy is achieved. Upon replacement of one Fe sublattice by V, the Heusler compound Fe₂VAl is realized, known to display bad-metal behavior and increased specific heat. We here document a charge-gap opening at low temperatures in line with previous experimental work. The gap structure does not match conventional band theory and is reminiscent of (pseudo)gap characteristics in correlated oxides.

I. INTRODUCTION

The Fe-Al system is well-known for its intricate phase diagram, displaying a complex interplay between metallicity, magnetism and structure. Stoichiometric FeAl poses a longstanding problem regarding its magnetic ground state. While experimentally B₂-FeAl is characterized as a Curie-Weiss paramagnet ¹¹ with no detectable ordered moment, conflicting results exist in theory.²³ On the Al-rich side, the low-symmetry structures Fe₃Al₂ and Fe₃Al₅ exhibit spin-glass physics at low temperature.⁰⁷ On the iron-rich side at Fe₃Al composition a bcc-based D₀₂₋ crystal structure is stable with well-defined ferromagnetic (fm) order up to Tᵣ = 713 K.⁵⁸ Further increase of the Fe content transforms the system into the doped bcc-Fe (or α-) phase, with fm order below a Curie temperature of 1043 K for pure iron. Albeit unambiguous in nature, both α-Fe and D₀₂₋-Fe₃Al appear difficult to be described within conventional density functional theory (DFT).⁹¹¹ The generalized-gradient approximation (GGA) for the exchange-correlation energy is necessary to detect the fm-bcc-Fe ground state.⁵⁰ Intriguingly, the fm-D₀₂₋ compound is only stable within the local-density approximation (LDA), while GGA favors the fcc-based L₁₂ structure in the ferromagnetic state.¹¹

This lack of coherent theoretical description of the Fe-rich side of Fe-Al in standard Kohn-Sham DFT asks for extended approaches. The inclusion of static electronic correlation effects via the DFT+Hubbard U method may cope with part of the subtle energetics for a reasonable choice of the local Coulomb-interaction parameters.¹² But that scheme is in principle not well defined for correlated itinerant systems and in addition usually needs to enforce magnetic order to deliver proper results. True paramagnetic states based on local-moment physics are neither accessible in conventional DFT nor in DFT+U.

A further facet of the intriguing correlated electronic structure in iron aluminides is revealed when replacing one Fe sublattice in D₀₂₋-Fe₃Al by vanadium. This transforms the intermetallic crystal to the Heusler L₁₂ compound Fe₂VAl. The ordered alloy is paramagnetic down to lowest temperatures and displays bad-metal behavior in transport.¹³ It is still a matter of debate if Fe₂VAl is a small-gap (~ 0.1 – 0.3 eV) semiconductor or a semimetal.¹⁴⁻¹⁵ Reminiscent of f-electron systems like SmB₆ with Kondo-insulating characteristics,¹⁶ heavy-fermion physics was originally associated with this 3d-electron system.¹⁷⁻¹⁸ Though magnetic defects later explained a sizable part of the large specific heat at low temperature, the overall mass enhancement remains substantial.¹⁹ Promising thermoelectric potential due to an enhanced thermopower is associated with Fe₂VAl-based materials.²⁰⁻²¹ Again, the theoretical first-principles assessment is difficult, since e.g. there are substantial differences concerning the existence of a charge gap Δ and its eventual size. Conventional DFT based on LDA/GGA classifies Fe₂VAl as semimetallic,²²⁻²³ whereas the use of hybrid functionals renders the system semiconducting with a band gap of Δₑ = 0.34 eV.²² A gap of Δₑ = 0.55 eV is revealed from DFT+U calculations.²³

In this work a first-principles many-body approach is employed to consider the effects of quantum fluctuations in the electronic structure of Fe₃Al and Fe₂VAl beyond conventional DFT(+U). The state-of-the-art combination of density functional theory with dynamical mean-field theory (DMFT) reveals important modifications to the correlated electronic structure. We show that the subtle electronic states rely on many-electron quantum processes, with important consequences for the phase stability and tendencies concerning gap formation. This paves the road towards a coherent modeling and understanding of Fe-Al and signals the general importance of advanced theoretical schemes for intermetallic systems.
FIG. 1. (color online) Relevant crystal structures. From left to right: D0_3-Fe_3Al, L1_2-Fe_3Al and L2_1-Fe_2VAl. Fe (red/light grey), Al (green/dark) and V (blue/grey).

II. CRYSTAL STRUCTURES

The crystal structures relevant for this work are displayed in Fig. 1. With bcc-Fe and fcc-Al as end members, the cubic lattice system also accounts for the dominant ordered phases in Fe-Al. Starting with B2-FeAl at stoichiometry, the bcc lattice is the common host for the stable solid phases in the Fe-rich part. Though the D0_3 structure is stable in the Fe_3Al phase regime, the fcc-based L1_2 structure appears as a relevant competitor. The D0_3 unit cell consists of three Fe and one Al site, whereas the Fe sites are grouped in two symmetry shells. One Fe site belongs to the Fe1 sublattice and two Fe sites to the Fe2 sublattice. As a bcc structure, each Fe site has 8 nearest-neighbor (NN) sites. Whereas the Fe2 atoms have mixed Fe/Al nearest neighborhood, the Fe1 atom has only Fe nearest neighbors. The experimental lattice constant of fully-ordered Fe_3Al reads a = 5.473 a.u.

The L1_2 structure also consists of three Fe and one Al atom in the primitive unit cell, but all Fe sites are equivalent by symmetry. The 12-atom NN shell of these Fe sites is composed again of both, Fe and Al sites. The measured lattice constant amounts to a' = 5.442 a.u.

Note that throughout this work we investigate the stoichiometric compounds, i.e. defect physics and effects of chemical disorder are not treated.

III. COMPUTATIONAL FRAMEWORK

The charge self-consistent DFT+DMFT methodology \cite{24,25} is put into practice, utilizing a mixed-basis pseudopotential approach\cite{26,27} for the DFT part and the continuous-time quantum-Monte-Carlo scheme\cite{28,29} from the TRIQS package\cite{30,31} for the DMFT impurity treatment. Exchange-correlation in the Kohn-Sham cycle is handled by the GGA functional of Perdew-Burke-Ernzerhof (PBE)\cite{32} form.

The correlated subspace where quantum fluctuations are explicitly accounted for is associated with the transition-metal sites of Fe and V kind. Projected-local orbitals\cite{33,34} of 3d character are used to extract Wannier-like states based on 22 Kohn-Sham bands, stemming from Fe/V(3d4s) and Al(3s3p) orbitals. Each transition-metal site represents a DMFT impurity problem, which due to symmetry amounts to two such ones in D0_3-Fe_3Al and Fe_2VAl, while only one symmetry-inequivalent transition-metal site is hosted in L1_2-Fe_3Al. A multi-orbital Hubbard Hamiltonian of Slater-Kanamori form, parametrized by the Hubbard U and the Hund’s exchange J_H, is applied to the respective full 5-orbital 3d-manifold. We overtook the values U = 3.36 eV and J_H = 0.71 eV for the local Coulomb interactions from Ref. 5 where those are computed for B2-FeAl using the constrained random-phase approximation. A double-counting correction of the fully-localized form is used in this work. If not stated otherwise, the temperature within the DMFT part is set to T = 387 K, i.e. \beta = 1/T = 30 eV^{-1}. The analytical continuation of the Green’s functions on the Matsubara axis i\omega is performed via the maximum-entropy method.

We restrict our DMFT calculations to phases without broken spin symmetry, i.e. no long-range ordered magnetism is modelled beyond DFT. Albeit D0_3-Fe_3Al is ferromagnetic at ambient temperatures, the magnetic ordering energy, as will be shown below, is not of vital importance for our investigation and its conclusions.

IV. RESULTS

A. Fe_3Al

1. Electronic states

We first focus in some detail on the electronic states in Fe_3Al. Let us start with the fcc-based L1_2 structure, having only one transition-metal sublattice. The close-packed lattice type is an important one in intermetallic...
systems, e.g. the ordered phases Cu₃Au and Ni₃Al condense in the L1₂ structure. Because of the cubic symmetry, here the local Fe(3d) states in principle split twofold into \( e_g \) and \( t_{2g} \) states. However due to the ordering pattern, not all \( e_g/t_{2g} \) sublevels may still be degenerate. This is illustrated in Fig. 2 where the obtained Fe(3d) projected local orbitals are plotted as isosurfaces. The \( e_g \) manifold consisting of \( \{ x^2-y^2, z^2 \} \) is defined by the orbital lobes pointing towards the next-nearest neighbor (NNN) Fe sites. Since both pointing directions are anisotropic in terms of the respective nearest-neighbor sites, the two \( e_g \) are non-degenerate. The \( t_{2g} \) manifold consisting of \( \{ xz, yz, xy \} \) are defined by the orbital lobes pointing to the NN sites. For \( xz, yz \) the associated NN sites are exclusively of Fe kind, therefore both orbitals are degenerate. Yet in the case of \( xy \) the associated NN shell consists exclusively of Al sites, thus this \( t_{2g} \) orbital has a different, in fact the lowest effective, crystal-field
Table I. Projected-local-orbital occupations in Fe$_3$Al. First lines are GGA, second lines DFT+DMFT results, respectively.

|          | $e_g$ | $t_{2g}$ | Total |
|----------|-------|----------|-------|
| $x^2-y^2$| 1.38  | 1.34     | 1.43  |
| $z^2$    | 1.55  | 1.56     | 1.79  |
| $xz$     | 1.43  | 1.54     | 1.79  |
| $yz$     | 1.43  | 1.54     | 1.79  |
| $xy$     | 1.58  | 1.54     | 1.79  |

**TABLE I. Projected-local-orbital occupations in Fe$_3$Al. First lines are GGA, second lines DFT+DMFT results, respectively.**

Figure 3 compares the integrated spectral functions $\rho(\omega) = \sum_k A(k, \omega)$ of L1$_2$-Fe$_3$Al within DFT(GGA) and DFT+DMFT. From the broadly itinerant band structure, an effective relevant bandwidth of about 7 eV (ranging from −6 eV to 1 eV) may be read off. Seemingly the full Fe(3$d$) manifold is crucial to understand the electronic structure in the bonding part and at low energy, since the hybridization between Fe and Al is rather strong in a wide energy range. Close to the Fermi level, the $z^2$ and $xy$ effective orbital are most dominant in GGA, while e.g. the $xz/yz$ part displays a bonding-antibonding nature.

For the $xy$ state with deepest crystal-field level and broad dispersion, the orbital filling is also largest (see Tab. I). The total local Fe electron count is slightly above seven within GGA. A further strengthening of the $xy$ dominance at low-energy occurs in the DFT+DMFT treatment. While the filling of all effective Fe orbitals increases with correlations, also here the occupation of the $xy$ state is enhanced largest by relative means. Overall, a substantial increase in the total effective Fe(3$d$) filling close to eight electrons takes place. Note that the site-filling differences between GGA and DFT+DMFT are also due to the respective effective-orbital definitions, as usual in determining local occupations in crystalline solids. First, the projected-local orbitals in both calculational schemes are not identical (only the projecting functions are), since via the charge self-consistent loop the Kohn-Sham part (i.e. the bands used for the projection) changes. Second, the resulting orbitals are of Wannier kind, i.e. their spread is not localized on the site centre within a small spherical radius.

Still correlations may enhance the electron localization on the Fe sites. The correlation strength can be estimated from the quasiparticle (QP) weight $Z \sim 1/m_{\text{eff}}$, computed from the electronic self-energy on the Matsubara axis as

$$Z = \left(1 - \frac{\partial \text{Im} \Sigma(i\omega)}{\partial \omega} \bigg|_{\omega \to 0^+}\right)^{-1}. \quad (1)$$

There is no strong orbital variation of the QP weight in the L1$_2$ structure and it amounts to a moderate value of $Z \sim 0.7$.

Though the D0$_3$ structure consists of two different Fe sublattices, the conventional internal degeneracies of the $e_g$ and $t_{2g}$ subshells of Fe(3$d$) are here fulfilled. This is due to the fact that the NN environments are either of pure Fe kind or of equally mixed-isotropic Fe,Al kind. As in fcc-based L1$_2$, the $e_g$ orbitals point again towards NN and NNN sites. However, since bcc-based D0$_3$ is not close packed, the $t_{2g}$ orbitals point interbetween the NN and NNN, i.e. towards the 3rd-nearest neighbor sites.

The total integrated spectral function of D0$_3$-Fe$_3$Al is similar to the one of L1$_2$-Fe$_3$Al (see Fig. 4), but with a more pronounced quasiparticle peak at low energy. The effective relevant bandwidth seems also smaller by about 1 eV in extent. On the local level, the Fe1 spectrum exhibits stronger $e_g$-$t_{2g}$ discrimination than the Fe2 spectrum. This speaks for a more subtle orbital/directional electronic structure around Fe1, whereas Fe2 with its 'washed-out' orbital signature looks similar to Fe in the
L1$_2$ structure. A strong GGA favoring of $e_g$ character at low energy in the case of Fe1 is weakened in DFT+DMFT, i.e. with correlations there are orbital-balancing tendencies at the Fermi level.

From the electron count, the Fe1($t_{2g}$) states become strongly correlation-enhanced, while on the other hand the Fe2($e_g$) electrons benefit from a local Coulomb interaction (cf. Tab. I). In principle, localizing D0$_3$ electrons in effective $t_{2g}$ orbitals is understandable from a charge-repulsion argument due to the orbital direction. Due to the stronger-hybridized environment on Fe2 imposed by nearby Al, there that single-site argument is not easily applicable. Note that the effective $e_g$ filling is levelled out in DFT+DMFT between Fe1 and Fe2. Figure underlines the present findings by inspecting the intra- and inter-site charge transfers. The bonding charge density $n^{\text{bond}} = n^{\text{crystal}}_n - n^{\text{atom}}$ with many-body effects shows furthermore charge depletion in the intersitial bonding region compared to the GGA result. In total, also the Fe sites in the D0$_3$ structure gain 3$d$ electrons upon the impact of local Coulomb interactions. While as expected the Fe2 site has a similar filling as the Fe site in L1$_2$, the Fe1 site has a lower electron count by roughly half an electron. Note that the absence of significant Fe filling differences with correlations in the recent work by Galler et al for B2-FeAl might be explained by the fact that no charge self-consistent framework was utilized in that study.

Concerning the correlation strength, though the Fe1 site and in general the $e_g$ orbital character has a somewhat lower QP weight, there is neither striking difference between the two Fe sublattices, nor between the $e_g/t_{2g}$ character. In numbers, an average value of $Z \sim 0.8$ is slightly higher than for the L1$_2$ structure, marking somewhat weaker correlation effects. The Fe$_3$Al compound is not a particularly strongly correlated material, since the ratio of the local Coulomb interaction and the bandwidth is well below unity. In addition, the local Fe occupation ranging between seven and eight electrons is already above the optimal Hund’s physics scenario of about 5 ± 1 electrons (where e.g. iron pnictides reside). Still correlation effects are effective in modifying the charge density and the low-energy character, having impact on bonding properties as well as on charge and spin response.

2. Energetics

We turn now to the structural phase competition between D0$_3$ and L1$_2$. In Fig. we compare the formation energy $E_{\text{form}}$ per atom with respect to the volume $V$, i.e.,

$$E_{\text{form}}^{\text{Fe$_3$Al}}(V) = E_{\text{tot}}^{\text{Fe$_3$Al}}(V) - \frac{3}{4} E_{\text{tot}}^{\text{bcc-Fe}}(V_{\text{eq}}) - \frac{1}{4} E_{\text{tot}}^{\text{bcc-Al}}(V_{\text{eq}}),$$

(2)

where $V_{\text{eq}}$ refers to respective equilibrium volume of the elemental phase. The data is based on nonmagnetic GGA and paramagnetic (pm) DFT+DMFT calculations. Both numerical schemes designate the D0$_3$ structure correctly as the stable one, with however two obvious differences. First, while in the many-body scheme the equilibrium volume is well reproduced, GGA yields a value too small by about 10%. Second, the energy difference $\Delta E_{D0_3}^{L1_2}$ between both structural types is about eight times larger within DFT+DMFT. Furthermore the bulk modulus $B$ is smaller in the latter scheme.

It was indeed shown in Ref. 11 that the first-principles description of the electronic structure and the phase stability of Fe$_3$Al is delicate. Upon ferromagnetic order, the L1$_2$ phase is by mistake stabilized in GGA(PBE). In this regard, a detailed data comparison is provided in Tab. 1. Disregarding explicit magnetic properties, while nm-GGA yields the correct qualitative structural hierarchy, the detailed structural data are off the experimental values. On the good side, introducing ferromagnetism on the GGA level brings lattice constant and bulk modulus close to experiment. However it not

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**TABLE II.** Comparison of structural data for D0$_3$-Fe$_3$Al. Formation energy $E_{\text{form}}$ (in meV/atom), bulk modulus $B$ (in GPa), lattice constant (in a.u.) and stability against the L1$_2$ structure. The (nm, fm, pm) formation energies are computed using the corresponding (nm, fm, pm) total energy of bcc-Fe.

|           | $E_{\text{form}}$ | $B$   | $a$ (Å) | stable |
|-----------|------------------|-------|--------|--------|
| nm-GGA    | -394             | 218   | 5.331  | ✓      |
| fm-GGA    | -202             | 151   | 5.465  |        |
| pm-DFT+DMFT | -325            | 143   | 5.480  | ✓      |
| experiment | -320 ± 20$^a$    | 144$^b$ | 5.473$^c$ | ✓      |

$^a$ Ref. 42, $^b$ Ref. 43, $^c$ Ref. 44
only misleadingly stabilizes the L12 structure but now strongly underestimates the formation energy. This major difference to the experimental $E_{\text{form}}^{\text{Fe}_3\text{Al}}$ appears not to solely linked to the GGA-type functional, but due to a general insufficient Kohn-Sham description of the magnetic energy in Fe-Al. Magnetism has been shown to be important for the D03 alloy ordering in that system. Also in the LDA-based work by Watson and Weinfert a value $E_{\text{form}}^{\text{Fe}_3\text{Al}} = -230$ meV/atom was obtained for spin-polarized D03-Fe3Al. From the computation of the formation energy of various Fe compounds, the authors there concluded that introducing spin polarization in the Kohn-Sham exchange-correlation functional underestimates the magnetic energy for such alloys.

The results of the DFT+DMFT scheme are overall in very good agreement with the available experimental data. Note that here in order to evaluate the formation energy, the bcc-Fe problem was of course also treated in paramagnetic DFT+DMFT with the identical local Coulomb interactions on Fe. The less negative $E_{\text{form}}$ of the D03 structure, in better agreement with experiment, matches the reduced bonding strength revealed from the correlated charge densities (cf. Fig. [5]). For the case of L12-Fe3Al, correlations not only render it much more energetically unfavorable compared to D03. Its formation energy becomes even positive, marking the compound unstable. This may be explained by the discussed correlation-induced weakening of the $xz/yz$ states with significant bonding-antibonding nature, compared to the strengthening of the $xy$ and $z^2$ states. Thus many-body effects beyond conventional DFT do not merely lead to some quantitative changes, but display a qualitative effect on the energetics in the Fe-Al system.

The general improvement in the theoretical description of D03-Fe3Al underlines not only the importance of electronic correlations, but renders it clear that a faithful description of the paramagnetic state is sufficient to account for the phase-relevant characterization.

B. Fe2VA1

In the final section, we discuss the electronic structure of the Heusler L21-Fe2VA1 compound that emerges from D03-Fe3Al by replacing the Fe1 sublattice with V atoms.

Figure 7 shows the total and local spectral properties, again by comparing GGA and DFT+DMFT. As in the earlier DFT-based studies, we find a semimetallic solution in the former Kohn-Sham calculation. A dichotomy is seen by inspecting the transition-metal electron-state characters on the local level. Below the Fermi level the low-energy region is dominated by Fe($t_{2g}$) states, while above $\varepsilon_F$ there are dominantly V($e_{g}$) states. As expected because of replacing the Fe1 ions, the V site has a more pronounced orbital differentiation. Yet due to the different vanadium valence, the GGA filling is of course only a bit more than half the size of the Fe site.

Within DFT+DMFT a clear gap structure emerges in the low-energy region, which is only fully realized at lower temperature. A pseudogap signature is obtained at a higher $T = 387$ K. It is notable that spectral weight is shifted towards the low-energy region to form sharp gap edges. Thus the gap formation is not of obvious single-particle character and has similarities with e.g. the (pseudo)gap structure in cuprates. Therefore the insulating state is not of conventional band-insulating semiconductor type. Measuring the charge-gap from the mids

![Figure 7](image_url)
of the gap-edge structure, a size $\Delta_g \sim 0.15\text{eV}$ is read off at $T = 193\text{K}$. This is in excellent agreement with experimental values for a charge gap in Fe$_3$VAI\cite{21}

Both transition-metal elements contribute to the intricate gap formation, but the V ion seems to play a more dominant role due to the larger spectral-function enhancement at the gap edges. Moreover the low-energy spectra with correlations displays are more balanced $e_g$,$t_{2g}$ contribution compared to GGA. This is in line with a nearly orbital-independent local self-energy on the V sites. Therefrom, the correlation strength is enhanced on the latter sites in comparison to the Fe sites, yet the vanadium-based QP weight $Z \sim 0.7$ is again moderate. Needless to say that Fe$_2$VAI is of course no Mott insulator. Still electronic correlations beyond conventional DFT are at the origin of the gap formation and -opening. Interesting in this context are the different electron fillings of V and Fe (see Tab. III). While the Fe ion non-surprisingly shows a very similar filling characteristic as the Fe2 ion in D0$_3$-Fe$_3$Al, the V ion already surely differs in the number of valence electrons. With an effective filling close to four electrons, the V site lies one hole below half filling, i.e. in a designated Hund’s metal regime.\cite{29,31}

The orbital-resolved V occupations align somewhat in DFT+DMFT, however it seems that the overall correlation strength due to the given sizes of bandwidth and local Coulomb interactions is yet too weak to drive very strong Hund’s physics. But unconventional spin fluctuations could nonetheless play a relevant role in the enhanced experimental specific heat.\cite{17}

|      | $e_g$  | $t_{2g}$ | $x^2 - y^2$ | $z^2$ | $xz$ | $yz$ | $xy$ | total |
|------|--------|----------|-------------|-------|------|------|------|-------|
| V    | 0.45   | 0.45     | 0.97        | 0.97  | 0.97 | 3.81 |
| Fe   | 0.70   | 0.70     | 0.90        | 0.90  | 0.90 | 4.10 |

TABLE III. Projected-local-orbital occupations in Fe$_3$VAI for $T = 387\text{K}$. First lines are GGA, second lines DFT+DMFT results, respectively.

V. CONCLUSIONS

Recently, there have been various investigations that employ realistic DMFT approaches beyond Kohn-Sham DFT(+U) to elemental iron and its alloy with aluminum. Studies on phase stabilities in the Fe phase diagram\cite{27,28} on the $\alpha$-Fe phonon spectrum\cite{33} on vacancy formation in $\alpha$-Fe\cite{34} and on the magnetism in B2-FeA\cite{35} were performed. The present work adds the DFT+DMFT examination of the Fe$_3$Al and Fe$_2$VAI correlated electronic structure.

We show that albeit both compounds do not fall in the standard category of strongly correlated systems, more subtle many-body effects are still relevant for a correct description. The energetics and phase stability of Fe$_3$Al builds upon such effects, by providing an improved value for the formation energy with a clear energy separation to the L1$_2$ structure. Note that the charge self-consistent version of the DFT+DMFT framework is important to elucidate such physics. Thereby not only local changes on the explicitly correlated lattice sites matter, but the overall charge redistribution including also interstitial and ligand region are crucial. On general grounds for cubic intermetallics, the open bcc lattice seems more adequate for correlated (Fe-based) compounds. For systems on the close-packed fcc lattice with sizeable local Coulomb interactions, the local correlations become comparatively too strong, weakening important bonding properties. Fcc-based compounds like e.g. Ni$_3$Al and Cu$_3$Au either do not display substantial local correlation physics or are well described in standard DFT.

The Fe$_2$VAI compound manifests an intriguing twist to traditional intermetallics. In the sense that the material is derived from the well-known Fe$_3$Al metal but displays an intricate gap opening reminiscent of (pseudo)gap physics observed in correlated oxides. The DFT+DMFT gap size and its sensitivity to temperature are in excellent agreement with experimental results for this compound. Since also Hund’s physics may play a role on the vanadium site, this example shows how easily traditional materials physics may be confronted with challenging mechanisms from strongly correlated matter. In a future work, addressing the thermoelectric properties of Fe$_3$VAI on the basis of the here established results would be highly interesting.

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