Comment on "Correlation Induced Paramagnetic Ground State in FeAl"

In contradiction with experiment [1], several band structure calculations [2–4] have shown FeAl in the CsCl(B2) structure to be ferromagnetic at least at the measured lattice parameter $a = 5.5$ a.u.

In a recent paper, Mohn *et al.* [5] use the LDA + U theory [6] to correct the local spin density approximation (LSDA) results and offer an explanation for the discrepancy between theory and experiment. In this work, the Hubbard U is used as an adjustable parameter and not a self-consistently calculated quantity. Therefore, in this case the LDA + U procedure is an empirical calculation to model the experiment and not a first-principles theory.

In this Comment, we wish to point out that an equivalent result can easily be obtained from a tight-binding (TB) Hamiltonian where the role of U is played by the Fe-t_{2g} on-site TB parameter.

A highly accurate TB Hamiltonian has been constructed to reproduce precisely the densities of states (DOS) derived from augmented plane wave (APW) calculations using the local density approximation (LDA). The details of the TB fit used here are described in Ref. [7]. The DOS values at the Fermi level reproduce the APW results to within 1%. We should emphasize that we fit only the energy bands in one structure. A more elaborate TB parametrization that is transferrable to other structures has not been used here.

The Stoner factor is $S = N(E_f)I$, where the exchange integral I was calculated using Janak's formalism [8]. In earlier work, we found that $S > 1$ for lattice parameters larger than 5.44 a.u. while for smaller lattice constants the Stoner criterion was not satisfied [3]. Here, we use the TB results at $a = 5.5$ a.u., the experimental value, and calculate S as the Fe-t_{2g} on-site parameter is reduced by Δ from its original value obtained in the fit. The LDA (and any TB parametrization to it) overestimates the energy of occupied localized orbitals [9]. As seen in Fig. 1, the Fe-t_{2g} states are nearly fully occupied, so the Fe-t_{2g} on-site parameter obtained from the fit should be higher than its "true" value. Decreasing the Fe-t_{2g} parameter effectively increases the t_{2g}-e_g crystal-field splitting. This decrease initially produces an increase of the Stoner factor S from 1.05 to 1.17 at a shift of the Fe-t_{2g} parameter by $\Delta = -0.74$ eV. However, for values of Δ less than -0.82 eV, S falls well below the critical value of 1 ($S = 0.63$ for $\Delta = -1.01$ eV) rendering FeAl paramagnetic.

The above result is absolutely equivalent to what was demonstrated by Mohn *et al.* [5] by varying U. To elaborate further on this point, the DOS for the unshifted TB parameters ($\Delta = 0$) and for $\Delta = -0.94$ are shown in Fig. 1. One can see that the main effect is the shift of the Fe-t_{2g} states to lower energies, reducing $N(E_f)$, the DOS at the Fermi level.

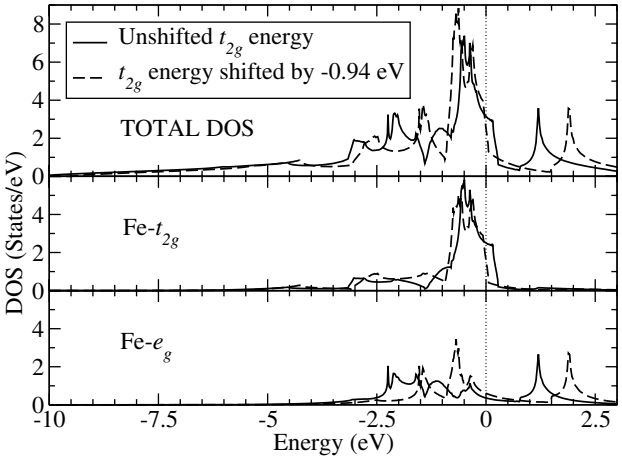

FIG. 1. Density of states (DOS) for the unshifted (solid lines) and shifted (dashed lines) TB parametrizations. The total DOS as well as the Fe-t_{2g} and Fe-e_g components are plotted.

In conclusion, we wish to reiterate the point that an accurate TB parametrization of the first-principles LDA band structure of FeAl can be used to produce a correction that brings about agreement with experiment regarding the absence of a ferromagnetic state. This is accomplished by adjusting the Fe-t_{2g} on-site parameter similarly to adjusting the Hubbard parameter U in the LDA + U theory.

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Received 2 January 2002; published 19 June 2002
DOI: 10.1103/PhysRevLett.89.029701
PACS numbers: 71.15.Mb, 75.20.En

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