Tight-binding v. LDA+U in FeAl

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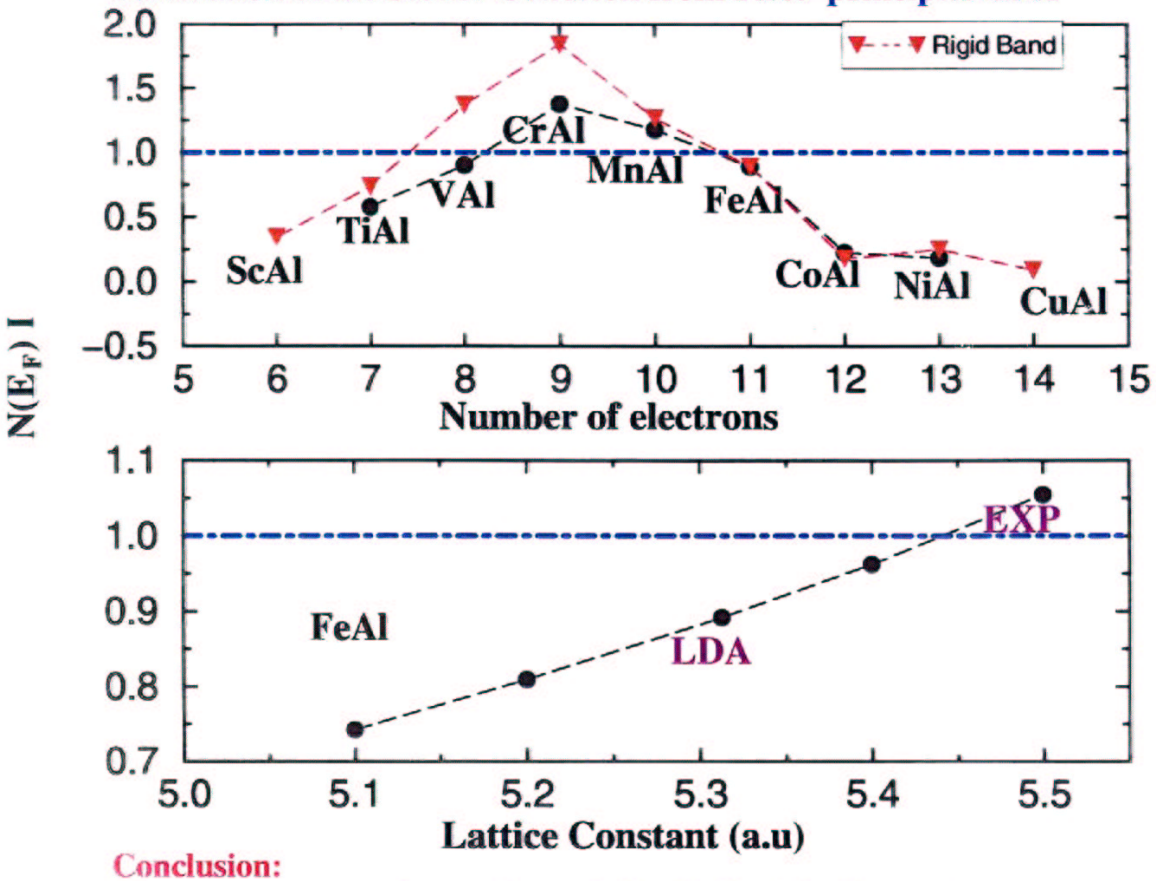
Search for Magnetism in Transition Metal Aluminides. B2 Structure.
Calculation of the Stoner Criterion from First–principles LDA



Conclusion:
We suggest an experimental search for V–Cr–Mn–Fe–Al alloys.

Stoner Criterion

A system will be ferromagnetic if

$$N(\varepsilon_F) I > 1$$

where

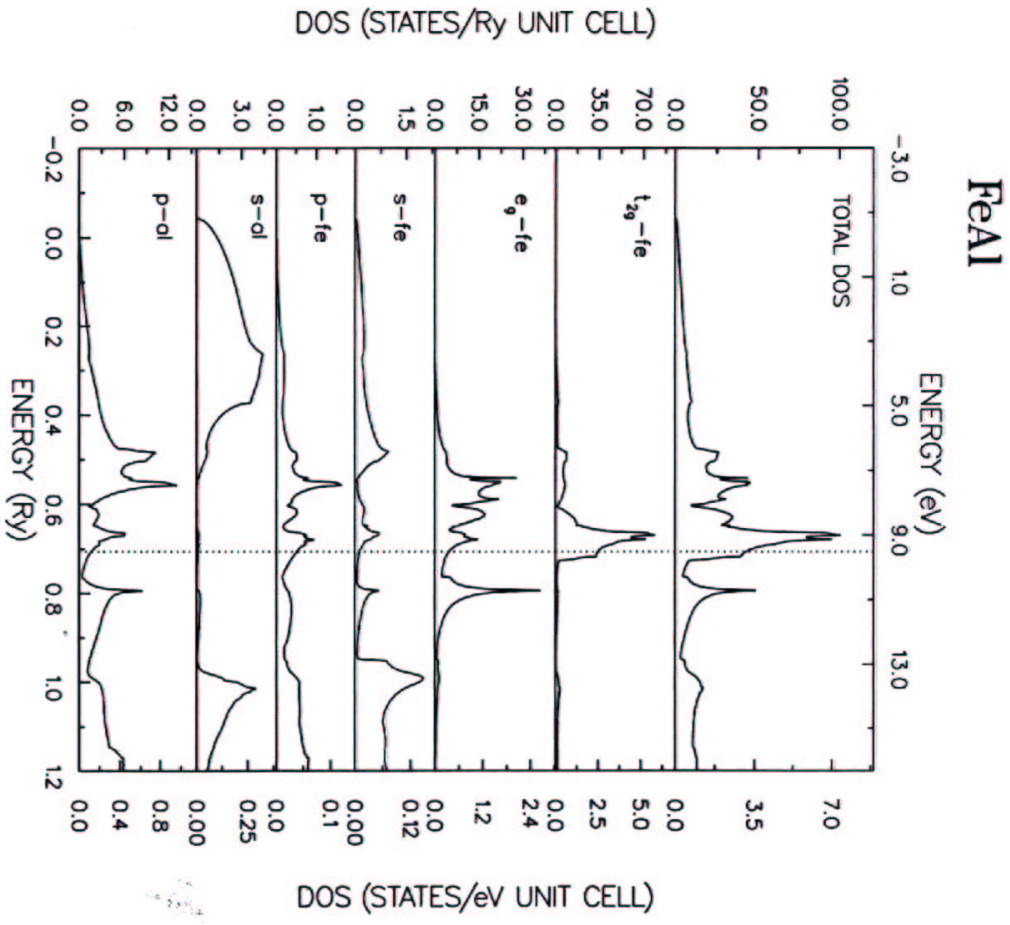
$$I = \int d^3r \, \gamma^2(\vec{r}) \, |K(\vec{r})| \ ,$$

$$\gamma = N(\varepsilon_F)^{-1} \sum_{\ell} N_\ell(\varepsilon_F) R_\ell^2(\varepsilon_F) \text{ and}$$

$$\left. \frac{\delta^2 E_{xc}[\rho; m]}{\delta m(\vec{r}) \delta m(\vec{r}')} \right|_{m=0} = 2K(\vec{r}) \delta(\vec{r} - \vec{r}') \ .$$

J.F. Janak, Phys. Rev. B16, 255 (1977)

S.H. Vosko and J.P. Perdew, Can. J. Phys. 53, 1385 (1975)



Mohn et al Phys. Rev. Lett. 87, 196401 (2001)

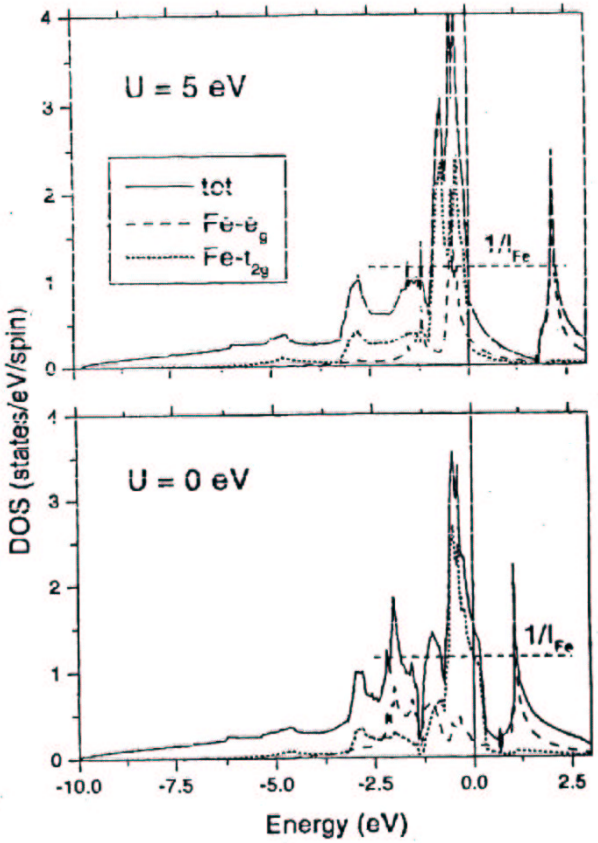
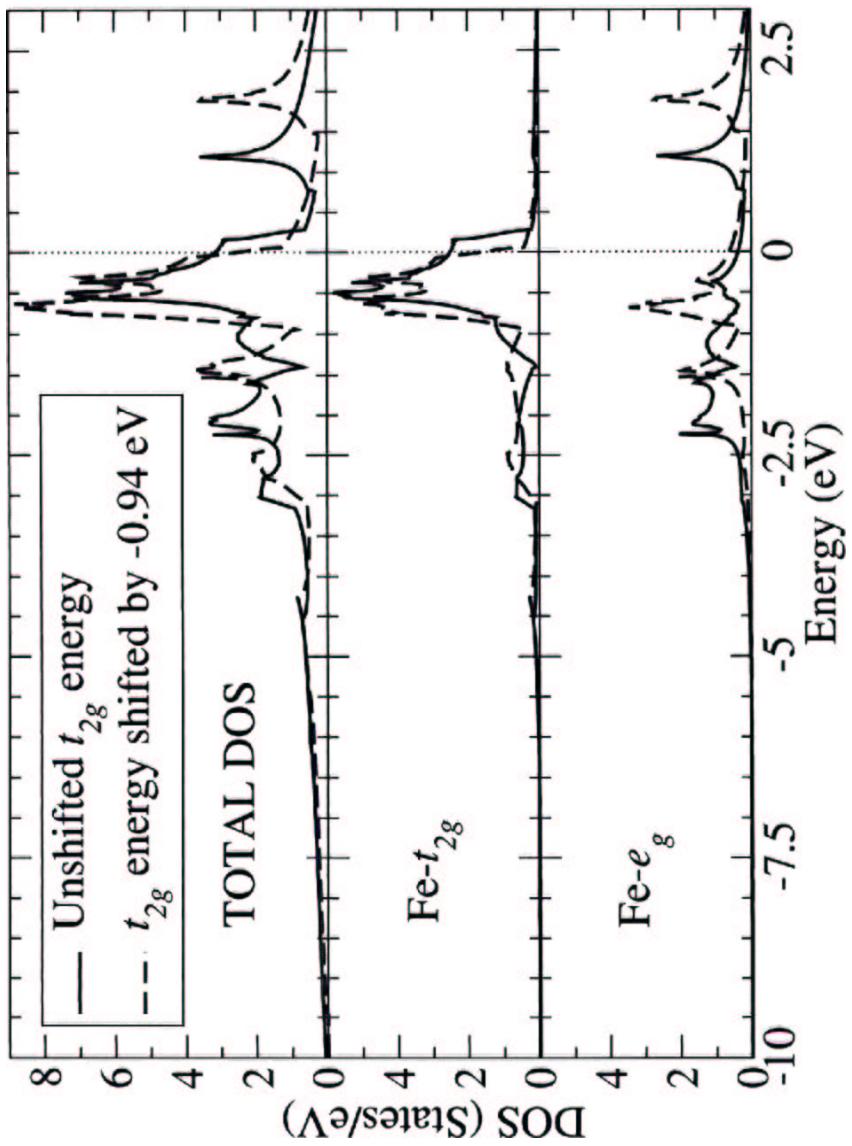


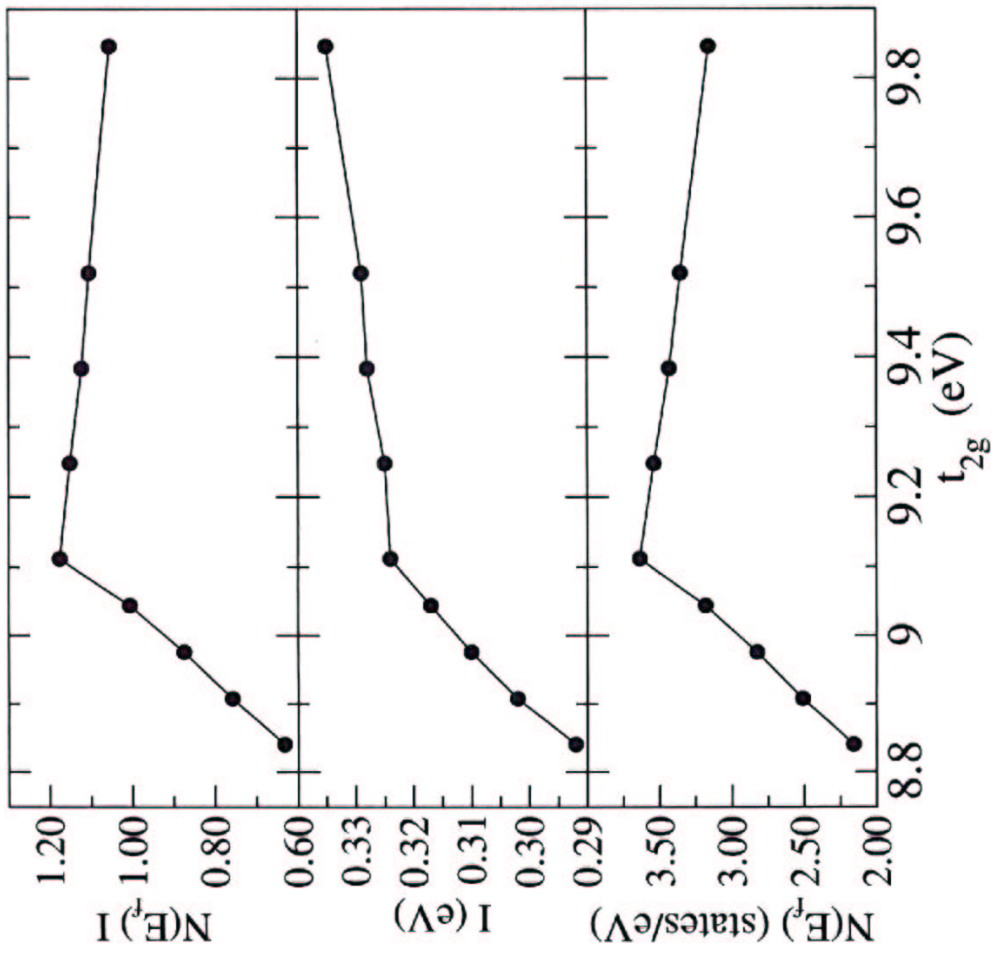
FIG. 1. Density of states (DOS) for $U = 5$ eV (upper panel) and $U = 0$ eV (lower panel) for FeAl and the decomposition into the Fe-e_g and Fe-t_{2g} manifolds for an LDA + U^{DPT} calculation in the nonmagnetic state. The horizontal dashed line marks the critical value for the DOS at ε_F above which the Stoner criterion ($1/I_{Fe}$) would be fulfilled.

Tight-binding Hamiltonian

1. Orthogonal s,p,d orbitals on both sites (18x18 secular equation)
2. 48 three-center parameters
3. Fit to LAPW calculations for 9 bands and 35 k-points
4. Hamiltonian block-diagonalized to account for symmetry
5. Fitting error less than 2 mRy
6. DOS calculated at 969 k-points by the tetrahedron method

Shore and Papa.. J. Phys. Chem. Solids 45, 439 (1984)

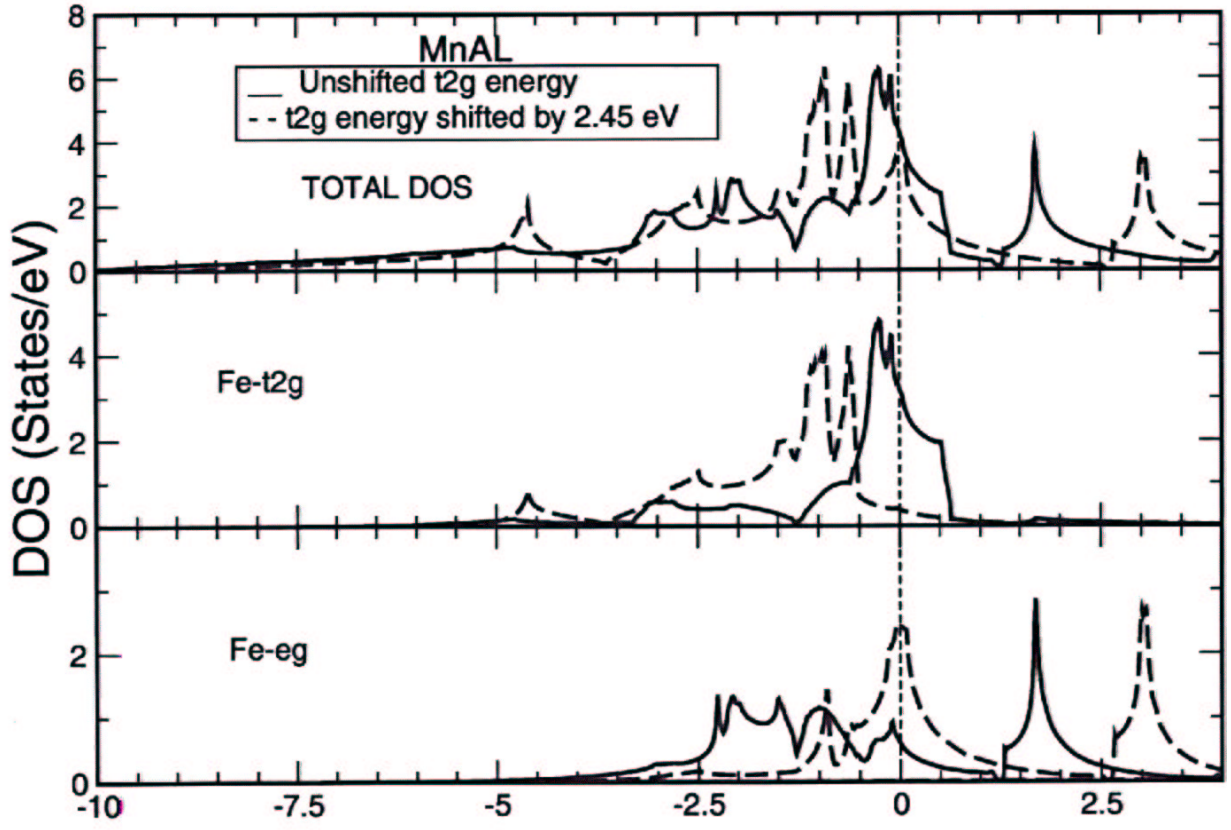

Unshifted t_{2g} energy
t_{2g} energy shifted by -0.94 eV
TOTAL DOS
Fe-t_{2g}
Fe-e_g
DOS (States/eV)
Energy (eV)

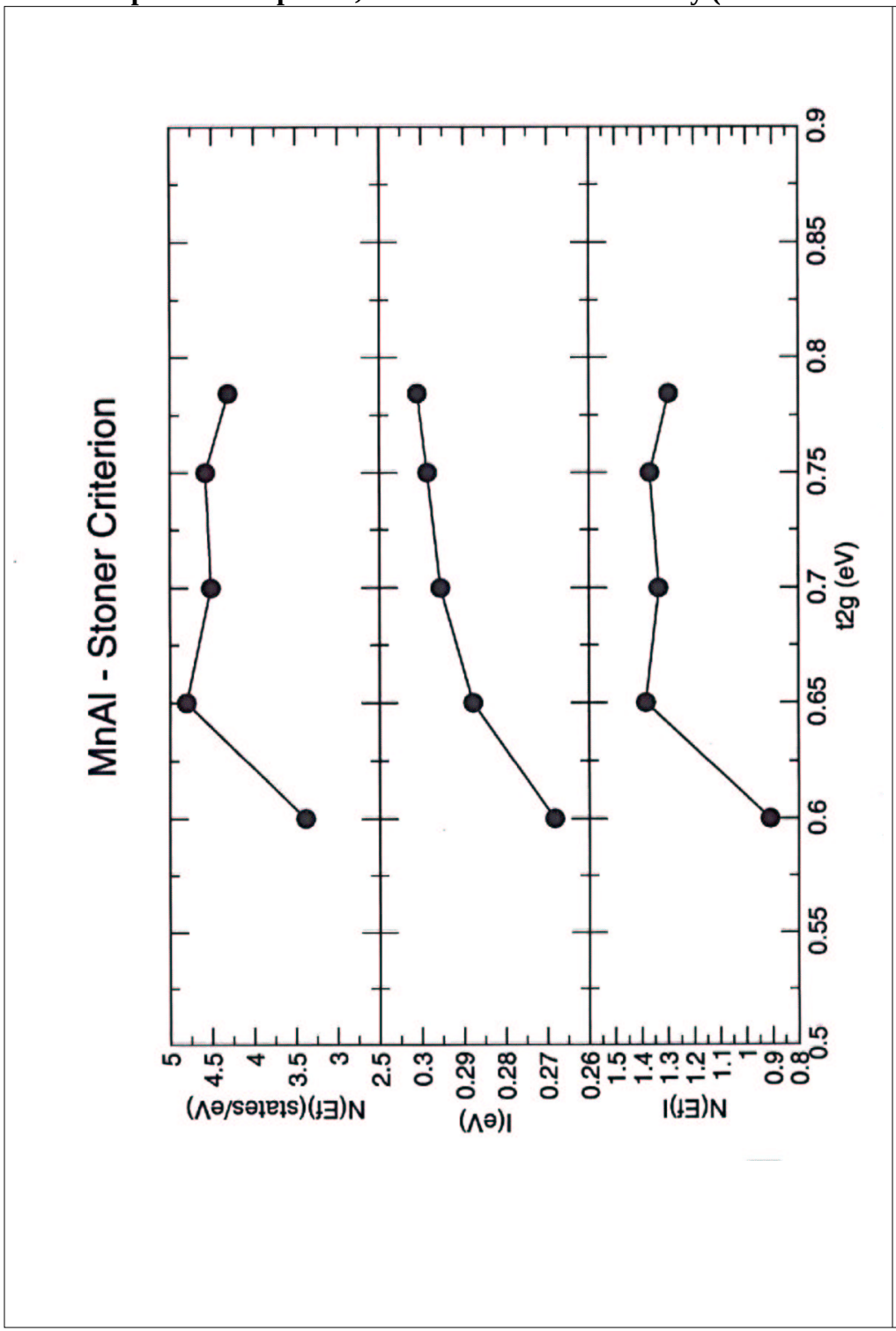

N(E$_F$) I
I (eV)
N(E$_F$) (states/eV)
t$_{2g}$ (eV)

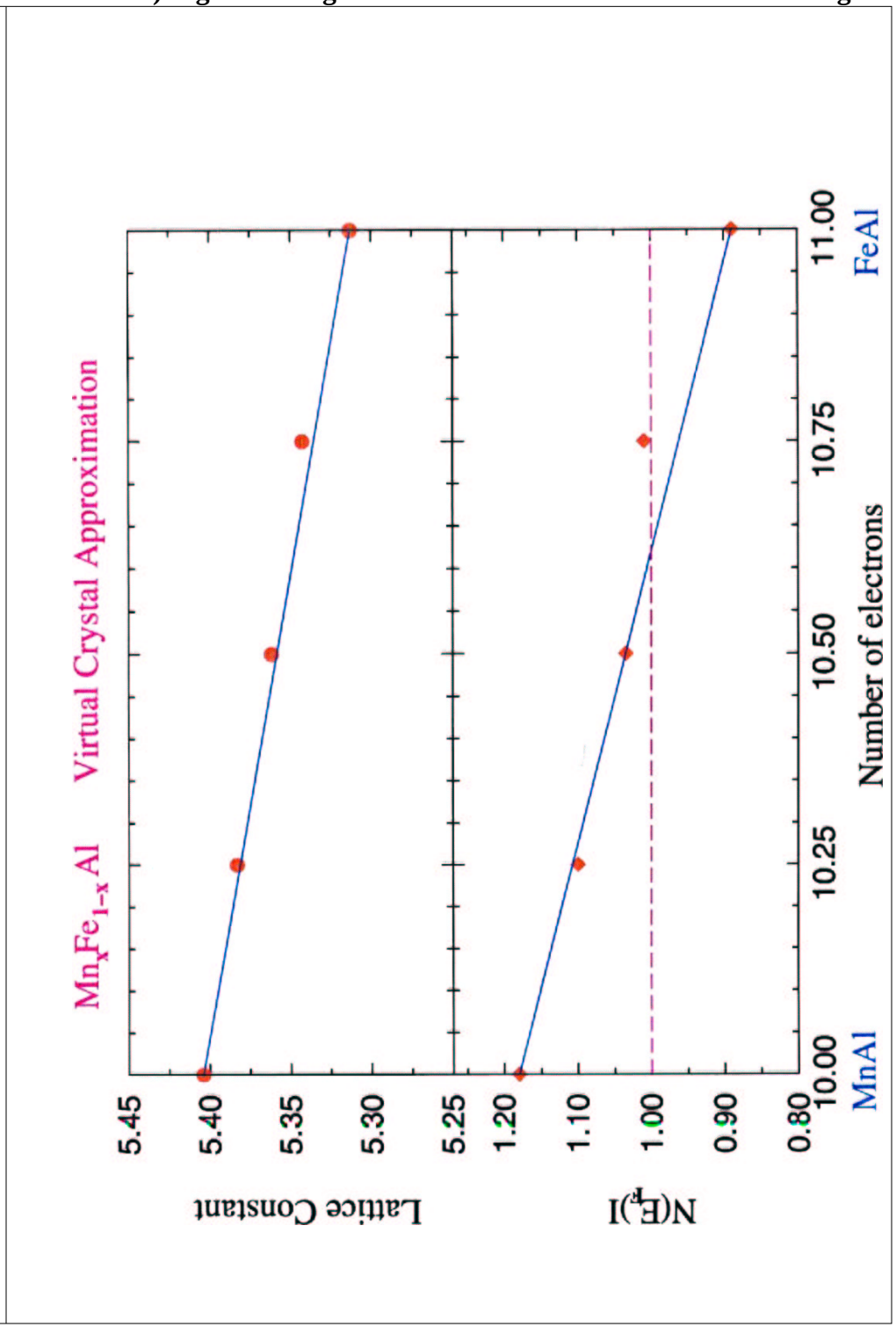
Integrated DOS

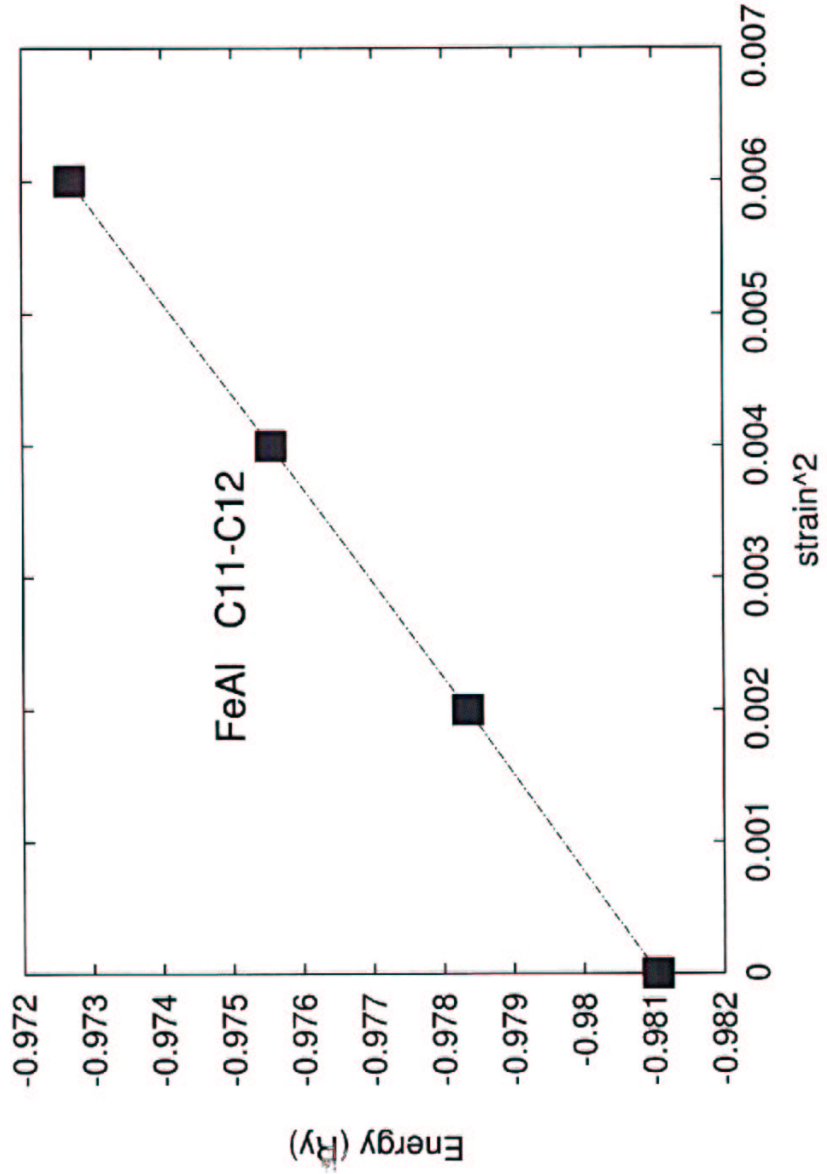
Original Total Elec	s-Fe	p-Fe	t2g-Fe	eg-Fe	s-Al	p-Al	t2g-Al	eg-Al
11.00	0.21	0.19	4.40	2.63	1.38	1.89	0.21	0.08

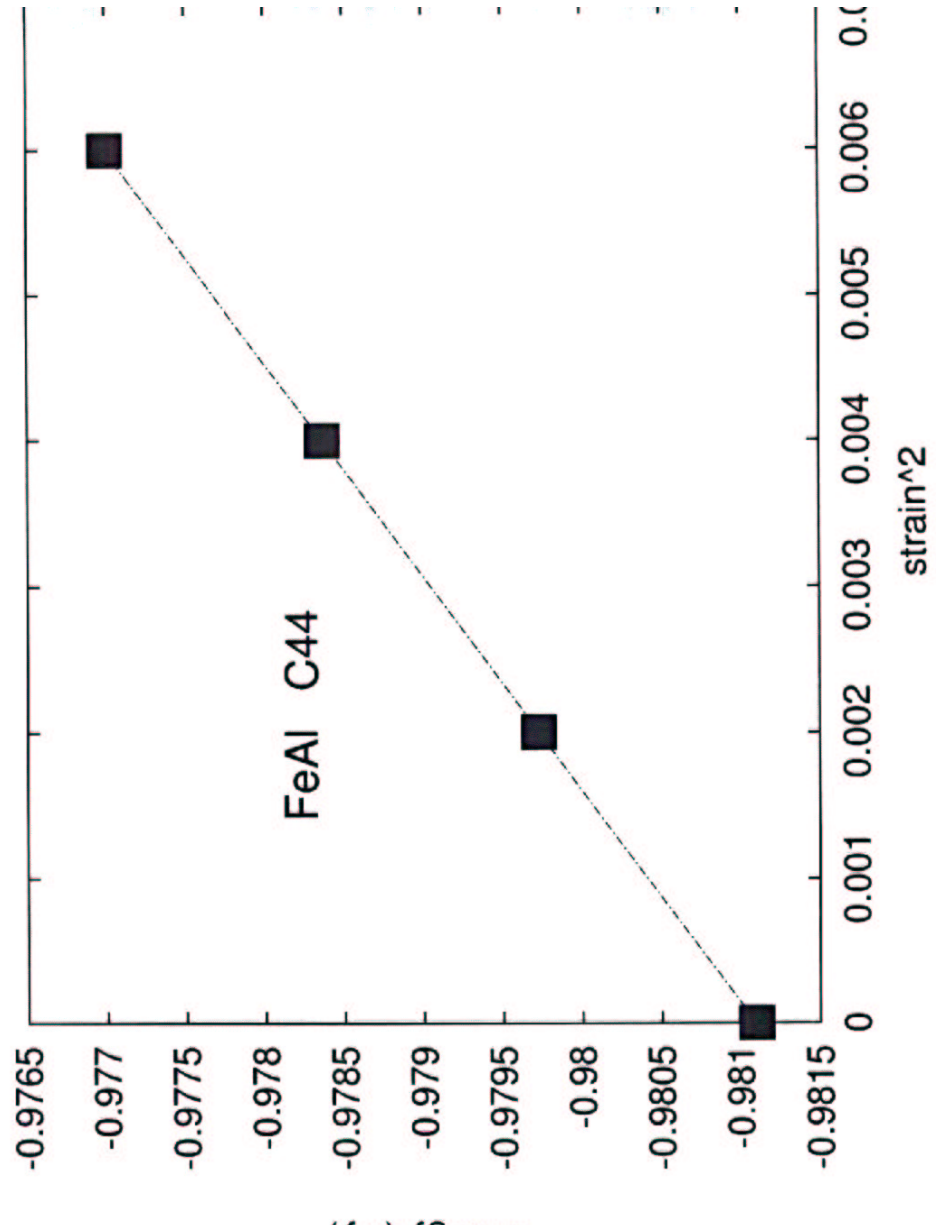
Shifted Total Elec	s-Fe	p-Fe	t2g-Fe	eg-Fe	s-Al	p-Al	t2g-Al	eg-Al
11.00	0.20	0.18	4.92	2.34	1.35	1.75	0.18	0.07

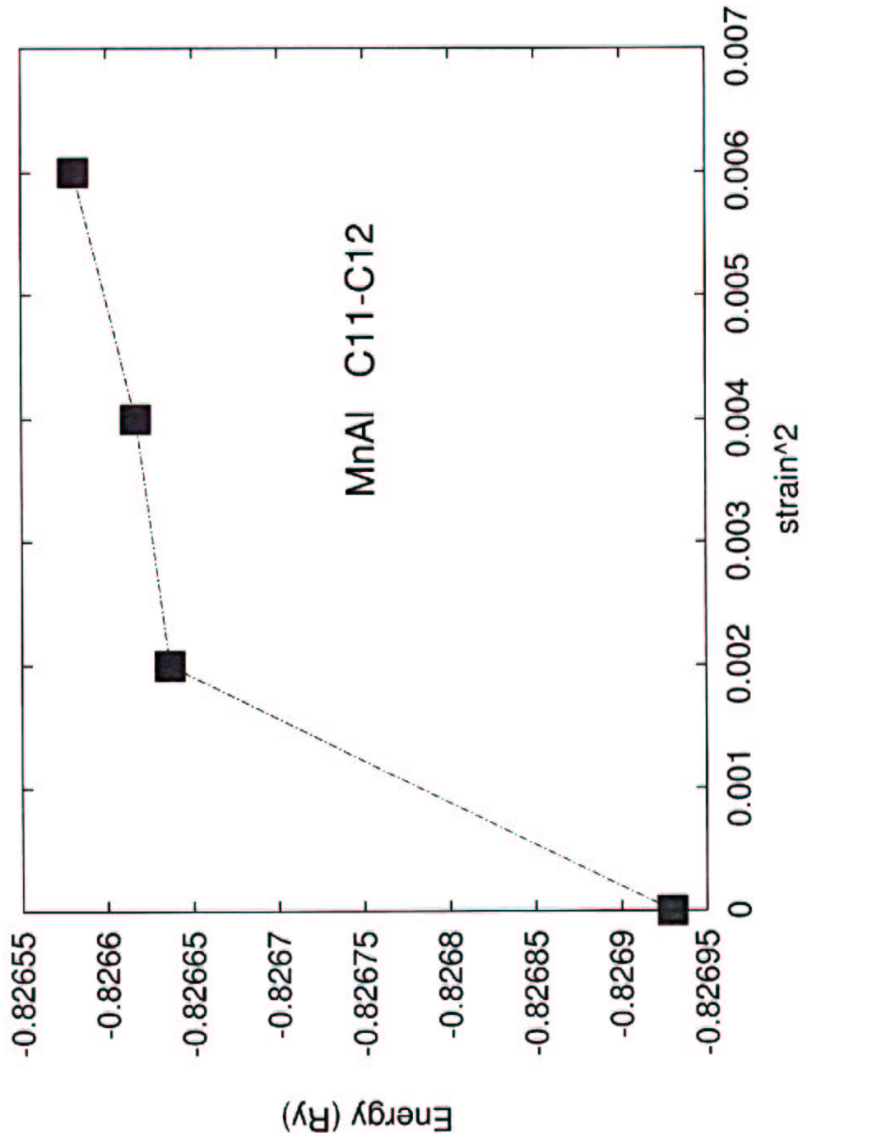


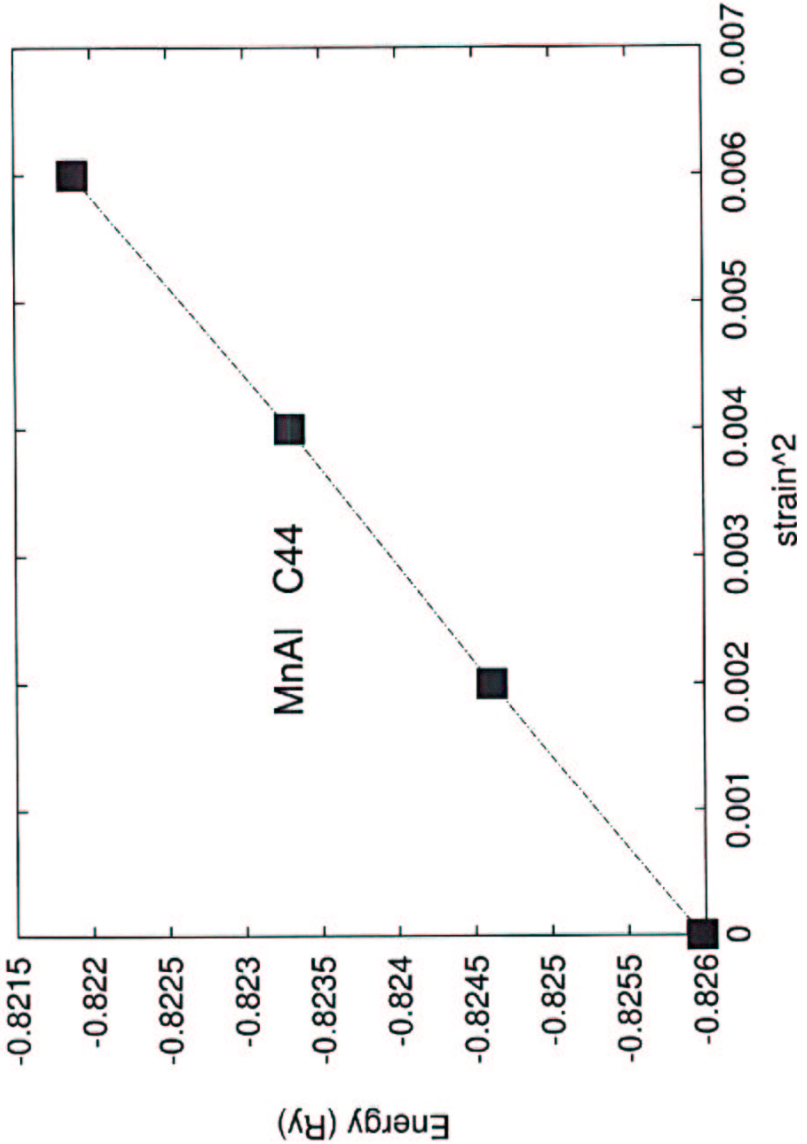

MnAl - Stoner Criterion
N(Ef)(states/eV)
I(eV)
N(Ef)I
t2g (eV)


$Mn_xFe_{1-x}Al$
Virtual Crystal Approximation
Lattice Constant
$N(E_F)I$
Number of electrons
MnAl
FeAl


FeAl C11-C12
Energy (Ry)
strain^2
-0.972
-0.973
-0.974
-0.975
-0.976
-0.977
-0.978
-0.979
-0.98
-0.981
-0.982
0
0.001
0.002
0.003
0.004
0.005
0.006
0.007


FeAl C44
Energy (Ry)
strain^2
-0.9765
-0.977
-0.9775
-0.978
-0.9785
-0.979
-0.9795
-0.98
-0.9805
-0.981
-0.9815
0
0.001
0.002
0.003
0.004
0.005
0.006


MnAl C11-C12
Energy (Ry)
strain^2
-0.82655
-0.8266
-0.82665
-0.8267
-0.82675
-0.8268
-0.82685
-0.8269
-0.82695
0
0.001
0.002
0.003
0.004
0.005
0.006
0.007


MnAl C44
Energy (Ry)
strain^2
-0.8215
-0.822
-0.8225
-0.823
-0.8235
-0.824
-0.8245
-0.825
-0.8255
-0.826
0
0.001
0.002
0.003
0.004
0.005
0.006
0.007

Conclusion

An accurate tight-binding parametrization of the LDA band structure of FeAl can be used to lower the Stoner criterion value below 1.

This is accomplished by adjusting the Fe-t_{2g} on-site TB parameter similarly to adjusting the Hubbard U in the LDA+U method. Both approaches basically increase the t_{2g}-e_g separation (crystal field splitting).