Magnetic instability in heavily *n*-doped Fe-based full-Heusler compounds for thermoelectric applications

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Using first-principles simulations, the Fe_2YZ full-Heusler compounds were predicted to exhibit very large thermoelectric power factor (PF), a central quality for thermoelectric applications [1]. Their essential feature is the simultaneous "flat-and-dispersive" character of the Fe e_g band [2], which can be engineered to increase PF by *n* doping [3]. However, the consequences of explicit *n* doping in these compounds have not been carefully investigated yet. Here, we study $Fe_2YZ_{1-x}A_x$ (Y=V, Ti, Ta, Nb, Z=Al, Si, Sn, Ga and A=Si, P, Sb, Ge) doped compounds with Density Functional Theory, using both Hybrid Functionals and GGA+U methods. We bring to light in some cases the appearance of a magnetic transition inducing a half-metal ferromagnetic phase. This behavior is shown to be intimately linked to the position of the Fe-e_g states with respect to the bottom of the conduction band, and to have a pure electronic origin, which is independent of the nature of dopants. In fact, by artificially varying the relative positions of the Fe-e_g and Y-e_g levels, we rationalize how the two regimes (non-magnetic or magnetic) are activated by electronic doping. Although the magnetic phase exhibits low PF, having a reduced number of carriers at the Fermi level with respect to the non-magnetic phase, it still remains promising for thermoelectric applications. We show, moreover, that the use of broader *d*-shells (4*d* or 5*d*), corresponding to much less localized electrons with respect to the 3*d* ones, pushes the onset of the magnetic transition to larger doping levels, and consequently allows recovering very large PF values.

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References

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[3] Phys. Rev. Lett. 114, 136601 (2015).