Supplementary Information

Highly spin-polarized electronic structure and magnetic properties of Mn2.25Co0.75Al1-xGex Heusler alloys: first-principles calculations

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A. Band structures of Mn_2CoAl and $Mn_{2.25}Co_{0.75}Al_{1-x}Ge_x$ at the ground states with SOC

The calculated band structures of Mn_2CoAl and $Mn_{2.25}Co_{0.75}Al_{1-x}Ge_x$ by considering SOC are shown in Fig. S1. It is found that SOC does not affect the HM/SGS characteristics of Mn_2CoAl and $Mn_{2.25}Co_{0.75}Al_{1-x}Ge_x$. Only some degenerated states are split into several singlet states at the high symmetry points.

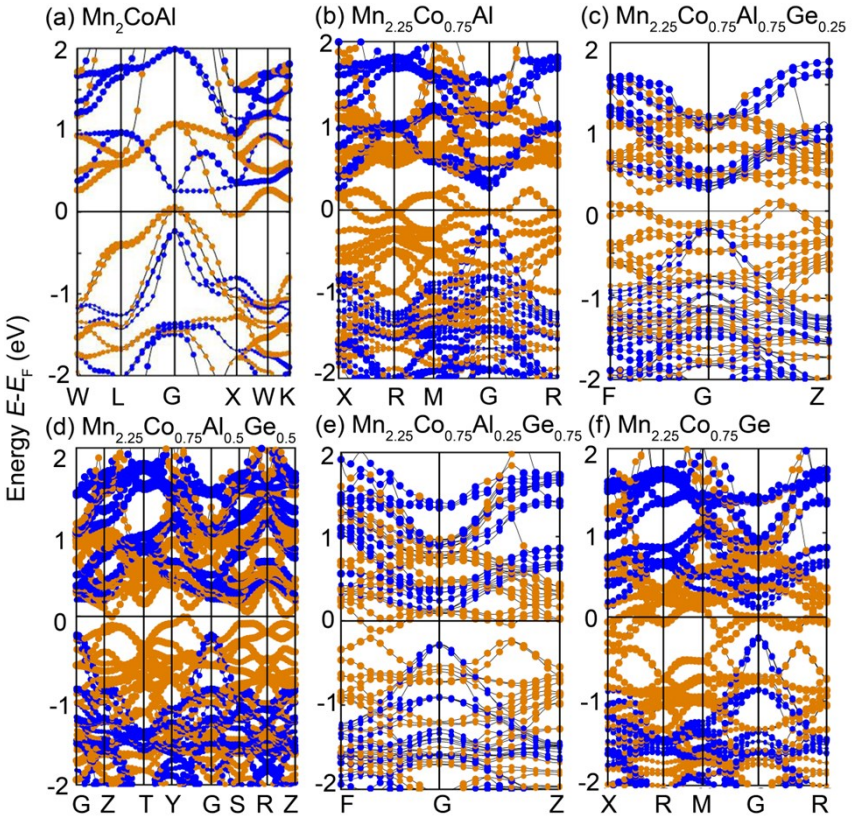

Fig. S1. Band structure of (a) Mn_2CoAl, (b) $Mn_{2.25}Co_{0.75}Al$, (c) $Mn_{2.25}Co_{0.75}Al_{0.75}Ge_{0.25}$, (d) $Mn_{2.25}Co_{0.75}Al_{0.5}Ge_{0.5}$, (e) $Mn_{2.25}Co_{0.75}Al_{0.25}Ge_{0.75}$ and (f) $Mn_{2.25}Co_{0.75}Ge$ at equilibrium lattice constant with SOC. The origin and blue circles represent the spin-up and spin-down channels, respectively.

B. Atomic-resolved MAE of Mn_2CoAl and $Mn_{2.25}Co_{0.75}Al_{1-x}Ge_x$ under uniaxial strains

The atomic-resolved MAEs of Mn_2CoAl and $Mn_{2.25}Co_{0.75}Al_{1-x}Ge_x$ at a uniaxial strain of -6%, 0% and 6% are shown in Fig.S2. In Fig.S2 (b)-(e), it can be clearly seen that the atomic-resolved MAEs of $Mn_{2.25}Co_{0.75}Al$, $Mn_{2.25}Co_{0.75}Al_{0.75}Ge_{0.25}$, $Mn_{2.25}Co_{0.75}Al_{0.5}Ge_{0.5}$ and $Mn_{2.25}Co_{0.75}Ge$ at a uniaxial strain of -6%, 0% and 6% display quite similar results with that of $Mn_{2.25}Co_{0.75}Al_{0.25}Ge_{0.75}$ which has been discussed detailly in the main work. The nearly zero MAE of the cubic $Mn_{2.25}Co_{0.75}Al$, $Mn_{2.25}Co_{0.75}Al_{0.75}Ge_{0.25}$, $Mn_{2.25}Co_{0.75}Al_{0.5}Ge_{0.5}$ and $Mn_{2.25}Co_{0.75}Ge$ (ε_t=0%) mainly comes from the minor opposite MAE of Mn(A2) atom in Ⅰ-layer and Mn(A3) atom in Ⅲ-layer. At ε_t=-6%, the positive MAE mainly comes from Mn(A2) and Co(C2) atom in Ⅰ-layer, Mn(C) and Co(C3) atoms in Ⅲ-layer. Mn(A3) atom in Ⅲ-layer contributes a relatively small negative MAE. Thus, $Mn_{2.25}Co_{0.75}Al_{1-x}Ge_x$ exhibits the in-plane magnetic anisotropy at ε_t=-6%. At ε_t=6%, every layer contributes a negative net MAE, where Co(C1), Mn(A3) and Mn(C) in Ⅰ-, Ⅲ-layers gives much contribution. Thus, the magnetic easy axis turns from the (100) direction into the (001) direction for $Mn_{2.25}Co_{0.75}Al$, $Mn_{2.25}Co_{0.75}Al_{0.75}Ge_{0.25}$, $Mn_{2.25}Co_{0.75}Al_{0.5}Ge_{0.5}$ and $Mn_{2.25}Co_{0.75}Ge$ with varying uniaxial strains. However, it should be noted that the origin of the MAE in Mn_2CoAl is different from that in $Mn_{2.25}Co_{0.75}Al_{1-x}Ge_x$ under uniaxial strain, as shown in Fig.S2 (a). The contribution of Mn(A) and Mn(B) atoms to MAE are negligible which is different from $Mn_{2.25}Co_{0.75}Al_{1-x}Ge_x$. And the negative and positive MAE at ε_t=6% and ε_t=-6% just origin from Co(C) atom.

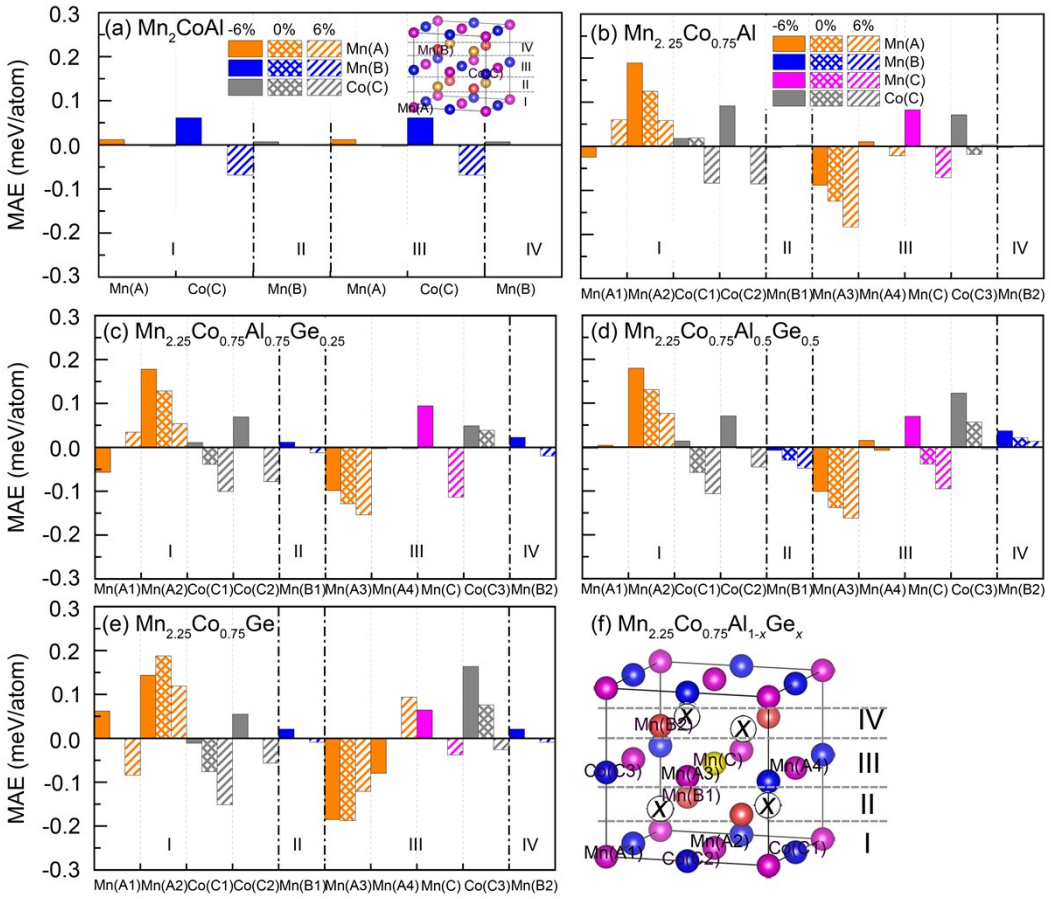

Fig. S2 Atomic-resolved MAE of (a) Mn_2CoAl, (b) $Mn_{2.25}Co_{0.75}Al$, (c) $Mn_{2.25}Co_{0.75}Al_{0.75}Ge_{0.25}$, (d) $Mn_{2.25}Co_{0.75}Al_{0.5}Ge_{0.5}$ and (e) $Mn_{2.25}Co_{0.75}Ge$ at a uniaxial strain of -6%, 0% and 6%. (f) The atomic structure of $Mn_{2.25}Co_{0.75}Al_{1-x}Ge_x$ (x=0, 0.25, 0.50, 1.00), where I-Ⅳ refers to the layer numbers. The inset of (a) is the atomic structure of Mn_2CoAl.