Supplementary information

Unusual Interfacial Magnetic Interactions for τ-MnAl with Fe (Co) Atomic Layers

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1. Computational details and models

For surface energy calculations of τ-MnAl, the symmetric τ-MnAl (001) slabs with Mn terminal or Al terminal are adopted. The slab is 15 atomic layers. The in plane lattice parameters are fixed and all of the atoms are allowed to relax in the out of plane direction.

For interface energy calculations of τ-MnAl/Fe (Co), the slabs are composed of both τ-MnAl and Fe (Co) are 11 atomic layers, respectively. The bottom four atomic layers of τ-MnAl are fixed to simulate the underlying bulk crystal. For the energy of Fe (Co) layers calculations, the in plane lattice parameters are fixed to match the lattice parameters of τ-MnAl and all of the atoms were allowed to relax in the out of plane direction. The magnetic states for $E_{E_{τ-MnAl/Fe(Co)}}$ (Eq.2) calculation were FM, AFM, FM and FM for Mn/Co, Mn/Fe, Al/Co and Al/Fe interface, respectively.

For τ-MnAl/Fe (Co) magnetic coupling calculations, the slabs are composed of 9 atomic layers τ-MnAl and different layers of Fe (Co). The bottom four atomic layers of τ-MnAl are fixed to simulate the underlying bulk crystal, as shown in Fig.1.

For the Fermi surface, cross section of Fermi surface and energy band calculations in Fig.8, Fig.10 and Fig.S3, the structures of bulk Fe and Co are convectional bct structure, which contain 2 atoms in one unit cell.
2. Results

Figure S1. Band structures of bct Co. The states for Co were projected onto (001) surface Brillouin zone. Red color for spin up bands (a) and blue color for spin down bands (b).
Figure S2. Spin up energy bands for FM coupling $\tau$-MnAl/Co with different layer numbers of Co. (a) 1 ML, (b) 2 ML, (c) 3 ML, (d) 4 ML, (e) 5 ML, (f) 6 ML, (g) 7 ML, (h) 8 ML and (i) 9 ML. The weightings of the Co atoms are projected on the band dispersions and marked by red circles.
Figure S3. Spin-resolved band structure of bct Fe. (a) Spin up band structure of bct Fe along ΓZ direction. The weightings for sum of \( s \), \( p_z \) and \( d_{z^2} \) orbitals the atoms are projected on the band dispersions and marked by red circles. Thus, the band marked by solid red circles is the spin up \( \Delta_1 \) band. (b) Spin down band structure of bct Fe along XR direction. The weightings of \( p \) orbital the atoms are projected on the band dispersions and marked by blue circles.
Figure S4. Spin up energy bands for FM coupling t-MnAl/Fe with different layer numbers of Fe. (a) 1 ML, (b) 2 ML, (c) 3 ML, (d) 4 ML, (e) 5 ML, (f) 6 ML, (g) 7 ML and (h) 8 ML. The weightings for the sum of Fe atoms $s$, $p_z$ and $d_{xz}$ orbital are projected on the band dispersions and marked by red circles.
Figure S5. Spin down energy bands for AFM coupling τ-MnAl/Fe with different layer numbers of Fe. (a) 1 ML, (b) 2 ML, (c) 3 ML, (d) 4 ML, (e) 5 ML, (f) 6 ML, (g) 7 ML and (h) 8 ML. The weightings of the Fe atoms are projected on the band dispersions and marked by blue circles.
Figure S6. Spin up energy bands for FM coupling r-MnAl/Fe with different layer numbers of Fe. (a) 1 ML, (b) 2 ML, (c) 3 ML, (d) 4 ML, (e) 5 ML, (f) 6 ML, (g) 7 ML and (h) 8 ML. The weightings of the Fe atoms are projected on the band dispersions and marked by red circles.
Figure S7. Spin up energy bands for AFM coupling τ-MnAl/Fe with different layer numbers of Fe. (a) 1 ML, (b) 2 ML, (c) 3 ML, (d) 4 ML, (e) 5 ML, (f) 6 ML, (g) 7 ML and (h) 8 ML. The weightings for $p$ orbital of Fe atoms are projected on the band dispersions and marked by red circles.
Figure S8. Spin down energy bands for FM coupling τ-MnAl/Fe with different layer numbers of Fe. (a) 1 ML, (b) 2 ML, (c) 3 ML, (d) 4 ML, (e) 5 ML, (f) 6 ML, (g) 7 ML and (h) 8 ML. The weightings for $p$ orbital of the Fe atoms are projected on the band dispersions and marked by blue circles.