SUPPLMENTAL MATERIAL FOR:

Phase stability and thermoelectric properties of semiconductor-like tetragonal FeAl2

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**Fig. S1.** The normalization of composition to comparing the Gibbs free energies at the same composition (Al 66.7at.%).



**Fig. S2.** The phonon band structures of (a) Fe3Al8-oP*44* and (b) its refined structure　Fe3Al8-*oP*44’. The imaginary phonon modes at gamma point were disappeared by shifting the atomic coordinates of Fe3Al8-*oP*44 in the direction of the eigenvector where the imaginary phonon modes appeared. The band-path was determined by using SeeK-path [1].

**Table S1**. The structural parameters of FeAl primitive cell obtained by the structural relaxation.

|  |  |  |  |
| --- | --- | --- | --- |
| Lattice parameters  |  |  |  |
| *a*(Å) | 2.875  |
| *b*(Å) | 2.875  |
| *c*(Å) | 2.875  |
| *α* | 90.000  |
| *β* | 90.000  |
| *γ* | 90.000  |
| Atomic coodinates | *x* | *y* | *z* |
| Fe1 | 0.500  | 0.500  | 0.500  |
| Al1 | 0.000  | 0.000  | 0.000  |

**Table S2.** The structural parameters of t-FeAl2 primitive cell obtained by the structural relaxation.

|  |  |  |  |
| --- | --- | --- | --- |
| Lattice parameters  |  |  |  |
| *a*(Å) | 3.003  |
| *b*(Å) | 3.003  |
| *c*(Å) | 4.750  |
| *α* | 71.572  |
| *β* | 71.572  |
| *γ* | 90.000  |
| Atomic coodinates | *x* | *y* | *z* |
| Fe1 | 0.000  | 0.000  | 0.000  |
| Al1 | 0.653  | 0.653  | 0.693  |
| Al2 | 0.347  | 0.347  | 0.307  |

**Table S3**. The structural parameters of a-FeAl2(FF) primitive cell obtained by the structural relaxation.

|  |  |  |  |
| --- | --- | --- | --- |
| Lattice parameters  |  |  |  |
| *a*(Å) | 4.803  |
| *b*(Å) | 6.344  |
| *c*(Å) | 8.785  |
| *α* | 87.523  |
| *β* | 74.498  |
| *γ* | 83.644  |
| Atomic coodinates | *x* | *y* | *z* |
| Fe1 | 0.143  | 0.155  | 0.418  |
| Fe2 | 0.857  | 0.845  | 0.582  |
| Fe3 | 0.220  | 0.345  | 0.888  |
| Fe4 | 0.780  | 0.655  | 0.112  |
| Fe5 | 0.000  | 0.000  | 0.000  |
| Fe6 | 0.166  | 0.478  | 0.583  |
| Fe7 | 0.834  | 0.522  | 0.417  |
| Al1 | 0.488  | 0.013  | 0.169  |
| Al2 | 0.512  | 0.987  | 0.831  |
| Al3 | 0.039  | 0.129  | 0.708  |
| Al4 | 0.961  | 0.871  | 0.292  |
| Al5 | 0.601  | 0.186  | 0.514  |
| Al6 | 0.399  | 0.814  | 0.486  |
| Al7 | 0.004  | 0.296  | 0.178  |
| Al8 | 0.996  | 0.704  | 0.822  |
| Al9 | 0.314  | 0.663  | 0.032  |
| Al10 | 0.686  | 0.337  | 0.968  |
| Al11 | 0.412  | 0.464  | 0.301  |
| Al12 | 0.588  | 0.536  | 0.699  |

**Table S4**. The structural parameters of a-FeAl2(AF) primitive cell obtained by the structural relaxation.

|  |  |  |  |
| --- | --- | --- | --- |
| Lattice parameters  |  |  |  |
| *a*(Å) | 4.866  |
| *b*(Å) | 6.464  |
| *c*(Å) | 8.636  |
| *α* | 88.728  |
| *β* | 74.392  |
| *γ* | 82.769  |
| Atomic coodinates | *x* | *y* | *z* |
| Fe1 | 0.137  | 0.165  | 0.416  |
| Fe2 | 0.854  | 0.839  | 0.582  |
| Fe3 | 0.233  | 0.360  | 0.866  |
| Fe4 | 0.772  | 0.637  | 0.137  |
| Fe5 | 0.989  | 0.000  | 0.995  |
| Fe6 | 0.168  | 0.474  | 0.594  |
| Al1 | 0.875  | 0.532  | 0.401  |
| Al2 | 0.493  | 0.994  | 0.162  |
| Al3 | 0.499  | 0.995  | 0.841  |
| Al4 | 0.037  | 0.116  | 0.710  |
| Al5 | 0.937  | 0.911  | 0.287  |
| Al6 | 0.589  | 0.182  | 0.541  |
| Al7 | 0.387  | 0.812  | 0.470  |
| Al8 | 0.045  | 0.286  | 0.152  |
| Al9 | 0.975  | 0.708  | 0.836  |
| Al10 | 0.300  | 0.663  | 0.041  |
| Al11 | 0.691  | 0.337  | 0.966  |
| Al12 | 0.446  | 0.424  | 0.299  |
| Al13 | 0.576  | 0.564  | 0.704  |

**Table S5**. The structural parameters of Fe3Al8-mC44 primitive cell obtained by the structural relaxation.

|  |  |  |  |
| --- | --- | --- | --- |
| lattice parameters  |  |  |  |
| *a*(Å) | 4.959  |
| *b*(Å) | 6.513  |
| *c*(Å) | 9.970  |
| *α* | 99.620  |
| *β* | 95.296  |
| *γ* | 97.322  |
| atomic coodinates | *x* | *y* | *z* |
| Fe1 | 0.341  | 0.147  | 0.199  |
| Fe2 | 0.678  | 0.881  | 0.814  |
| Fe3 | 0.659  | 0.534  | 0.140  |
| Fe4 | 0.330  | 0.484  | 0.859  |
| Fe5 | 0.670  | 0.198  | 0.480  |
| Fe6 | 0.322  | 0.801  | 0.526  |
| Al1 | 0.976  | 0.978  | 0.035  |
| Al2 | 0.029  | 0.478  | 0.552  |
| Al3 | 0.971  | 0.204  | 0.787  |
| Al4 | 0.024  | 0.704  | 0.303  |
| Al5 | 0.158  | 0.427  | 0.086  |
| Al6 | 0.833  | 0.574  | 0.912  |
| Al7 | 0.167  | 0.108  | 0.428  |
| Al8 | 0.830  | 0.886  | 0.567  |
| Al9 | 0.450  | 0.145  | 0.691  |
| Al10 | 0.550  | 0.873  | 0.304  |
| Al11 | 0.450  | 0.808  | 0.035  |
| Al12 | 0.526  | 0.198  | 0.972  |
| Al13 | 0.474  | 0.484  | 0.367  |
| Al14 | 0.550  | 0.537  | 0.649  |
| Al15 | 0.842  | 0.254  | 0.253  |
| Al16 | 0.170  | 0.796  | 0.772  |

**Table S6.** The structural parameters of Fe3Al8-oP44 and Fe3Al8-oP44’ primitive cells obtained by the structural relaxation. The atomic coordinates of Al21, Al22, Al23, and Al24 were mainly shifted in the Fe3Al8-oP44’.

|  |  |
| --- | --- |
| Fe3Al8-*oP*44 | Fe3Al8-*oP*44' |
| lattice parameters  |  |  |  |  |  |  |  |
| *a*(Å) | 12.401  |  | 12.394  |
| *b*(Å) | 7.617  |  | 7.619  |
| *c*(Å) | 6.392  |  | 6.393  |
| *α* | 90.000  |  | 90.000  |
| *β* | 90.000  |  | 90.000  |
| *γ* | 90.000  |  | 90.000  |
| atomic coodinates | *x* | *y* | *z* |  | *x* | *y* | *z* |
| Fe1 | 0.089  | 0.250  | 0.076  |  | 0.078  | 0.249  | 0.085  |
| Fe2 | 0.415  | 0.750  | 0.583  |  | 0.578  | 0.249  | 0.415  |
| Fe3 | 0.915  | 0.750  | 0.917  |  | 0.405  | 0.749  | 0.573  |
| Fe4 | 0.589  | 0.250  | 0.424  |  | 0.905  | 0.749  | 0.927  |
| Fe5 | 0.243  | 0.250  | 0.427  |  | 0.240  | 0.249  | 0.425  |
| Fe6 | 0.256  | 0.750  | 0.926  |  | 0.740  | 0.249  | 0.075  |
| Fe7 | 0.756  | 0.750  | 0.574  |  | 0.252  | 0.749  | 0.926  |
| Fe8 | 0.743  | 0.250  | 0.073  |  | 0.752  | 0.749  | 0.574  |
| Fe9 | 0.418  | 0.250  | 0.078  |  | 0.418  | 0.249  | 0.074  |
| Fe10 | 0.918  | 0.250  | 0.422  |  | 0.918  | 0.249  | 0.426  |
| Fe11 | 0.077  | 0.750  | 0.576  |  | 0.079  | 0.749  | 0.580  |
| Fe12 | 0.577  | 0.750  | 0.924  |  | 0.579  | 0.749  | 0.920  |
| Al1 | 0.062  | 0.250  | 0.712  |  | 0.082  | 0.249  | 0.715  |
| Al2 | 0.562  | 0.250  | 0.788  |  | 0.582  | 0.249  | 0.785  |
| Al3 | 0.419  | 0.750  | 0.214  |  | 0.445  | 0.749  | 0.215  |
| Al4 | 0.919  | 0.750  | 0.286  |  | 0.945  | 0.749  | 0.285  |
| Al5 | 0.078  | 0.054  | 0.377  |  | 0.080  | 0.048  | 0.377  |
| Al6 | 0.578  | 0.054  | 0.123  |  | 0.580  | 0.048  | 0.123  |
| Al7 | 0.418  | 0.549  | 0.876  |  | 0.418  | 0.557  | 0.880  |
| Al8 | 0.918  | 0.549  | 0.624  |  | 0.918  | 0.557  | 0.620  |
| Al9 | 0.418  | 0.951  | 0.876  |  | 0.418  | 0.942  | 0.880  |
| Al10 | 0.918  | 0.951  | 0.624  |  | 0.918  | 0.942  | 0.620  |
| Al11 | 0.578  | 0.446  | 0.123  |  | 0.580  | 0.451  | 0.123  |
| Al12 | 0.078  | 0.446  | 0.377  |  | 0.080  | 0.451  | 0.377  |
| Al13 | 0.255  | 0.059  | 0.118  |  | 0.249  | 0.071  | 0.089  |
| Al14 | 0.755  | 0.059  | 0.382  |  | 0.749  | 0.071  | 0.411  |
| Al15 | 0.245  | 0.569  | 0.597  |  | 0.240  | 0.555  | 0.623  |
| Al16 | 0.745  | 0.569  | 0.903  |  | 0.740  | 0.555  | 0.877  |
| Al17 | 0.245  | 0.931  | 0.597  |  | 0.240  | 0.944  | 0.623  |
| Al18 | 0.745  | 0.931  | 0.903  |  | 0.740  | 0.944  | 0.877  |
| Al19 | 0.755  | 0.441  | 0.382  |  | 0.749  | 0.428  | 0.411  |
| Al20 | 0.255  | 0.441  | 0.118  |  | 0.249  | 0.428  | 0.089  |
| Al21 | 0.290  | 0.250  | 0.780  |  | 0.335  | 0.249  | 0.745  |
| Al22 | 0.790  | 0.250  | 0.720  |  | 0.835  | 0.249  | 0.755  |
| Al23 | 0.174  | 0.750  | 0.256  |  | 0.218  | 0.749  | 0.288  |
| Al24 | 0.674  | 0.750  | 0.244  |  | 0.718  | 0.749  | 0.212  |
| Al25 | 0.416  | 0.076  | 0.427  |  | 0.408  | 0.069  | 0.404  |
| Al26 | 0.916  | 0.076  | 0.073  |  | 0.908  | 0.069  | 0.096  |
| Al27 | 0.086  | 0.572  | 0.912  |  | 0.078  | 0.576  | 0.929  |
| Al28 | 0.586  | 0.572  | 0.588  |  | 0.578  | 0.576  | 0.571  |
| Al29 | 0.086  | 0.928  | 0.912  |  | 0.078  | 0.923  | 0.929  |
| Al30 | 0.586  | 0.928  | 0.588  |  | 0.578  | 0.923  | 0.571  |
| Al31 | 0.916  | 0.424  | 0.073  |  | 0.908  | 0.430  | 0.096  |
| Al32 | 0.416  | 0.424  | 0.427  |  | 0.408  | 0.430  | 0.404  |

**Reference**

[1] Hinuma Y, Pizzi G, Kumagai Y, Oba F and Tanaka I Band structure diagram paths based on crystallography Comput. Mater. Sci. 2017 ;128 :140–84