Journal of Magnetism and Magnetic Materials ∎ (∎∎∎∎) ∎∎∎–∎∎∎

Contents lists available at ScienceDirect

Journal of Magnetism and Magnetic Materials

journal homepage: www.elsevier.com/locate/jmmm

First-principle prediction of robust half-metallic Te-based half-Heusler alloys

Shi-Yuan Lin [a], Xiao-Bao Yang [a], Yu-Jun Zhao [a,b,*]

[a] Department of Physics, South China University of Technology, Guangzhou 510640, China
[b] State Key Laboratory of Luminescent Materials and Devices, South China University of Technology, Guangzhou 510640, China

ARTICLE INFO

Article history:
Received 4 June 2013
Received in revised form
20 August 2013

Keywords:
First-principle calculation
Half-Heusler alloy
Half-metal

ABSTRACT

Te-based half-Heusler systems are studied by first-principle calculations to search for alloys with stable half-metallic properties. We found that CoMnTe and FeMnTe are the most robust half-metallic (HM) ferromagnetic alloys among the 90 studied alloys, with HM gaps of 0.42 and 0.61 eV, respectively, larger than that of any Heusler or half-Heusler alloys reported in the literature. The half-metallicity of CoMnTe and FeMnTe is found to be robust under large in-plane strains, which makes them suitable for practical spintronic device applications.

1. Introduction

Half-metallic (HM) ferromagnets have attracted much interest in the past decades since their 100% spin polarized states at the Fermi level hold great promise for applications in spintronics [1]. So far HM property has been reported in magnetic oxides [2–5], double perovskite [6], rare-earth nitrides [7,8], zinc-blende transition metal pnictides [9–11], chalcogenides [12], and Heusler alloys [13–20]. The HM Heusler alloys have relatively high Curie temperature [21,22] compared with other HM systems, and their structures match well with zinc-blende structure (including the similar diamond structure), which dominates the semiconductors electronic industry.

In addition to the stability of ferromagnetism, the magnitude of the HM gap (the energy difference between the highest occupied spin-up and spin-down states) is crucial for the application of half-metal Heusler alloys, since it affects the performance of spin injection devices. Unfortunately, most of the reported HM half-Heusler alloys have small HM gaps. For instance, the first-principle calculations predicted HM gaps for FeMnSb [23], NiCrP [24], NiCrSe [24], NiCrTe [24], and NiVAs [25] are only 0.2, 0.263, 0.047, 0.102, and 0.07 eV, respectively. The small HM gaps would often disappear in these half-Heusler alloys when strains from mismatch exist in their interface with conventional semiconductors. Therefore, exploring new HM materials with large HM gaps is crucial for practical spintronic device applications, although half-Heusler alloys CoCrP and CoCrAs were predicted to be HM ferromagnetic alloys with much larger HM gaps (~0.5 eV) recently [26].

In this work, we performed systematic search of the Te-based half-Heusler alloys through first-principle calculations, and found that CoMnTe and FeMnTe are robust HM alloys with large HM gaps of 0.42 and 0.61 eV and band gaps in minority spin channel of 1.13 and 1.24 eV, respectively. The half-metallicity of CoMnTe and FeMnTe can be maintained even under in-plane strains of −11.3% to 6.1% and −11.7% to 10.0%, respectively.

The calculations were carried out with the spin polarized density functional theory (DFT) as implemented in Vienna *ab initio* simulation package (VASP) [27]. The generalized gradient approximation (GGA) of PBE [28] was adopted for the exchange and correlation functional. A plane wave cutoff of 450 eV and a $5\times5\times5$ Monkhorst–Pack k-point mesh are used for the calculation. The total energy and eigenvalues are converged to be within 10 meV and the HM band gap changes within 3 meV with respect to the k-points according to the test calculations with a k mesh of $9\times9\times9$ for most of the alloys with HM gaps of >0.2 eV, including CoVTe, FeVTe, CoCrTe, FeCrTe, CoMnTe, and FeMnTe. We also considered the site preference of X and Y atoms in XYTe. We found that the atoms with larger number of valence electrons prefer X site for most of the systems, except NiCoTe, Cu based and Zn based systems. In particular, most of the interesting alloys with HM gaps larger than 0.2 eV, including CoVTe, FeVTe, CoCrTe, FeCrTe, CoMnTe and FeMnTe, obey the valence electron rule and their energy differences for site preference are more than 500 meV/atom in general. This means that these large gap half-metal systems are rather robust with respect to site preference. Nevertheless, we will discuss all the possible half-metallic properties of XYTe with both X and Y from Sc to Zn. For the calculations

* Corresponding author at: South China University of Technology, Department of Physics, 381 Wushan Road, Guangzhou 510640, China. Tel.: +86 20 87110426; fax: +86 20 87112837.
E-mail address: zhaoyj@scut.edu.cn (Y.-J. Zhao).

0304-8853/$ - see front matter © 2013 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.jmmm.2013.09.026

of local magnetic moments and projected density of states, the default PAW radii from VASP code are used. To confirm the reliability of our calculation, we also calculated the HM gaps of CoCrP and CoCrAs. The calculated HM gaps are 0.42 and 0.51 eV, correspondingly, which are in excellent agreement with the FLAPW values obtained by Yao et al. [26] (0.46 and 0.51 eV, respectively).

2. XYTe half-Heusler alloys

The crystal structure of half-Heusler compounds is described by the space group $F\bar{4}3m$ with $C1_b$ structure and the atomic arrangement is presented in Fig. 1. The positions of the basis atoms in Wyckoff coordinates are as follows: X atoms at (0 0 0), Y atoms at (1/4 1/4 1/4) and Te atoms at (3/4 3/4 3/4), in analogy with filled tetrahedral compounds [29].

In order to explore systems with larger HM gaps, various Te-based half-Heusler alloys XYTe (X=Sc–Zn, and Y=Sc–Zn) are investigated. It was reported that some Mn containing half-Heusler compounds may not be stable in contrast to other structures. For instance, CoMnSb crystallizes in a cubic superstructure, which can be illustrated by alternating MnSb and Co_2MnSb cells [30]. These half-Heusler compounds, however, may be still stabilized by strains, which are hard to avoid in the interface of devices. The strain effect on the HM properties will be addressed later. Here, we consider the stability of ferromagnetic (FM) and antiferromagnetic (AFM) phases in a 12-atom unit cell. Of note, here AFM and FM configurations refer to the spin arrangement of element Y in XYTe since Y contributes the most to magnetic moments. In detail, half of the Y atoms in a tetrahedron were aligned antiparallel to the other half. We found that 49 alloys of the studied 90 candidates are in favor of FM phases. Most of the alloys with FM phases are in the region of X=V–Co, but only a few of them follow the Slater–Pauling rule [31] with integral total magnetic moment. On the other hand, 41 alloys are in AFM configurations which correspond to alloys of X=Ni–Zn, or X=Sc–Ti, except Y=Mn–Co. For instance, CuCrTe favors an AFM configuration, with the local magnetic moments of Cr atoms around 4.07 μ_B. We also found that most of the alloys with FM phases contain atoms with large magnetic moment, such as Cr, Mn, and Fe, while the AFM alloys often contain atoms with negligible local moment, such as Sc, Cu, and Zn.

All the calculated band gaps and HM gaps are shown in Fig. 2 for Te-based half-Heusler alloys XYTe (X=Sc–Zn, and Y=Sc–Zn with $X \neq Y$). It is clear that the systems with large band-gap are mostly in the middle region, which corresponds to X=Cr–Co, Y=Ti–Mn. Clearly the HM gaps are much smaller, with only a few alloys having relatively large HM gaps, which correspond to alloys of X=Fe or Co, Y=V–Mn.

In Table 1, we list the calculated equilibrium lattice constant, energy difference between the AFM and FM phases, minority spin channel band gap, and HM gap for most of the alloys with HM gaps larger than 0.2 eV, including CoVTe, FeVTe, CoCrTe, FeCrTe, CoMnTe, and FeMnTe. It is clear that their FM phases are rather stable with respect to the corresponding AFM phases. In particular, the exchange energies for CoCrTe, FeCrTe, CoMnTe, and FeMnTe are all greater than 100 meV/formula. We found that the HM gap of FeMnTe is as large as 0.61 eV, larger than any reported HM gaps for Heusler or half-Heusler alloys in the literature. Its corresponding band gap in minority spin channel reaches 1.24 eV, indicating that the Fermi level is pinned nearly at the mid-gap to maximize the HM gap. In the

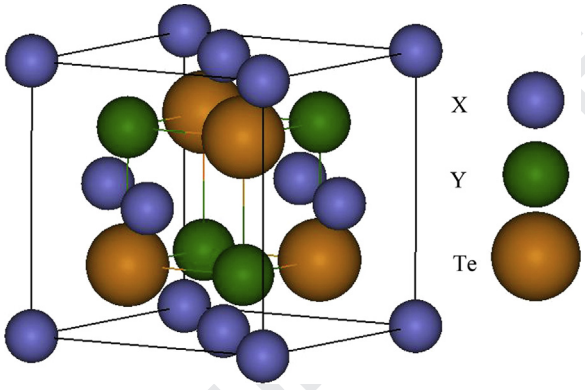

Fig. 1. The crystal structure of half-Heusler compounds XYTe.

Table 1
Calculated equilibrium lattice constant (a_0), energy difference between the antiferromagnetic and ferromagnetic phases (ΔE), band gap (E_g), and HM gap (E_g^{HM}) of the HM half-Heusler alloys XYTe. Here, AFM and FM configurations refer to the spin arrangement of element Y in XYTe.

Compound	a_0 (Å)	ΔE (eV/formula)	E_g (eV)	E_g^{HM} (eV)
CoVTe	5.88	0.09	1.05	0.21
FeVTe	5.82	0.06	1.31	0.20
CoCrTe	5.87	0.12	0.78	0.27
FeCrTe	5.87	0.13	0.91	0.34
CoMnTe	5.86	0.17	1.13	0.42
FeMnTe	5.84	0.12	1.24	0.61

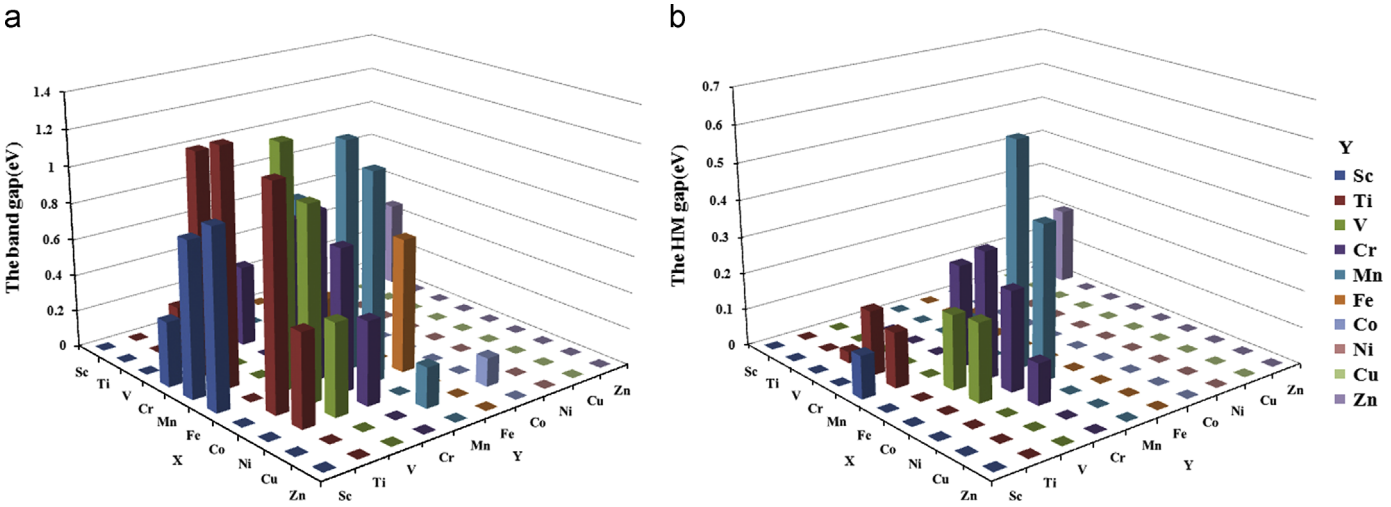

Fig. 2. The band gaps (a) of minority spin channel and corresponding HM gaps (b) of Te-based half-Heusler XYTe alloys.

Please cite this article as: S.-Y. Lin, et al., Journal of Magnetism and Magnetic Materials (2013), http://dx.doi.org/10.1016/j.jmmm.2013.09.026