



## Half-metallicity and ferromagnetism of TcX (X=C, Si and Ge) in zinc blende structure

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### ABSTRACT

We report results of a first-principles density-functional study of three binary transition-metal compounds TcX (X=C, Si and Ge) in the hypothetical cubic zinc blende (ZB) structure. Our calculations are based on the full potential linear augmented plane wave (FP-LAPW) plus local orbitals method, together with generalized gradient approximation for the exchange-correlation potential. Half-metallic (HM) ferromagnetism is observed in these binary compounds for their optimized cell volumes. In the HM state, these compounds possess an integer magnetic moment ( $1.000\mu_B$ ) per formula unit, which is one of the important characteristics of half-metallic ferromagnets (HMFs). The ferromagnetic (FM) state is found to be stable for ZB TcC, TcSi and TcGe against the nonmagnetic (NM) and antiferromagnetic (AFM) states. Calculations show that half-metallicity can be maintained for a wide range of lattice constants in these binary compounds. Density functional calculations of exchange interactions and the Curie temperatures reveal similar trends for the three compounds with respect to the lattice parameter. These compounds are compatible with the traditional semiconductors, and could be useful in spin-electronics and other applications. The most important aspect of this work is to explore the possibility of not only magnetism, but HM ferromagnetism in compounds involving NM elements and 4d transition element Tc.

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### 1. Introduction

In recent years, a great deal of effort has been devoted to study the HM materials in view of their importance in the newly developing field of spin-based electronics or spintronics [1–3]. In 1983, de Groot et al., via density functional calculations, reported the existence of HM ferromagnetism in magnetic Heusler compounds such as NiMnSb [4]. HMFs are of immense interest because of 100% electron spin-polarization at the Fermi level. These materials are semiconductors with energy gaps for one spin direction, and metallic for electrons of opposite spin. Thus the electrons at the Fermi level, responsible for transport properties, are all spin-aligned. Subsequent to the study by de Groot et al., there have been numerous theoretical predictions and experimental studies on the HM materials such as NiMnSb, CrO<sub>2</sub>, Fe<sub>3</sub>O<sub>4</sub>, manganite materials showing colossal magnetoresistance, and some transition-metal chalcogenides and pnictides [5–14]. Moreover, ZB Mn-IV binary compounds were also found to be HMFs [15–17].

The need for finding new HMFs which are more promising in basic properties and for applications [18] still remains paramount. The aim is to search for half-metals with simple structure, and high Curie temperature. Recently Liu et al. [19] and Bose and Kudrnovský [20] have presented extensive studies of exchange interactions and the Curie temperatures as a function of lattice parameter for some Cr-based chalcogenides and pnictides exhibiting half-metallicity. HM compounds which do not involve transition metals have also been considered. ZB Ca-based pnictides were predicted to be HMFs by Kusakabe et al. [21]. Very recently the HM ferromagnetism was found in many I–IV, I–V, II–IV and II–V ZB compounds [22–26], such as NaC and CaAs, where no transition metals are involved.

Almost all transition metal-based studies of half-metallicity involve elements from the 3d series. Though there are some works related to HM compounds containing no transition metal elements, those based on 4d transition elements are notably absent. The existence of the 4d transition element technetium was predicted by Mendeleev, who noted a gap in the periodic table indicating the missing element. The element was later discovered by Perrier and Segre in Italy in 1937. Nearly all technetium is produced synthetically and only minute amounts are found in nature. Technetium was the first element not found in nature (in any significant amount) to

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be produced artificially. It is a silvery-gray metal that tarnishes slowly in moist air, and its chemistry is supposed to be similar to that of its 5d counterpart, rhenium. Technetium's most stable isotopes, technetium-97 and technetium-98, have half-lives of 2.6 million years and 4.2 million years, respectively. Small amounts of technetium can retard the corrosion of steel. Technetium is used in a wide variety of diagnostic tools in medicine [27]. Until 1960s technetium was available only in small amounts. It can now be made in much larger quantities. Increase in technetium production is bound to follow as new applications of technetium and its compounds emerge. This work is a step in that direction.

Electronic properties of technetium are interesting. It has the second highest superconducting transition temperature among the elemental solids, surpassed marginally by niobium. Experiments as well as theoretical calculations reveal a high electron–phonon coupling [28]. Calculations for its close-packed hexagonal phase show that it has a moderately high Coulomb pseudopotential, moderate to high phonon frequencies and relatively low electron–spin fluctuation (paramagnon) coupling [28]. The latter is consistent with the observation that the exchange-enhancement of its spin susceptibility [29] is similar to its 4d neighbors, Ru and Rh, and relatively low compared to some of the 3d transition metals. However, under optimum conditions it can be driven toward magnetism. Magnetic properties of the transition metals in general, and especially the trends, can be understood to a large extent by noting that the *d*-band width decreases from left to right along a particular row and increases from 3d to 4d, and then to 5d along a particular column. Narrowness of a *d*-band results in a high density of states at the Fermi level, which drives the system toward Stoner instability and encourages lowering of energy via spin-polarization, i.e. magnetism. Wider *d*-bands are hard to spin-polarize, hence the absence of magnetism in the 4d counterparts of Mn, Fe, Co and Ni, namely Tc, Ru, Rh and Pd. However, the possibility of rendering these 4d elements, particularly Ru and Rh, magnetic via lattice dilation and/or reduced co-ordination, has been discussed in the literature and claims of experimental observation of such 4d magnetism have been made [30–32]. It should thus be possible to induce the same effect in Tc.

One way to put Tc atoms in an under-coordinated environment and thus render it magnetic would be to consider Tc-based alloys, where the hybridization between Tc 4d-orbitals is drastically reduced. Tc-based alloys in open (i.e. not close-packed) Zinc blende structure provide this opportunity. The binary transition-metal compounds TcX (X=C, Si and Ge) are structurally compatible with the group IV semiconductors, and share the zinc blende crystal structure with the typical semiconductor materials such as GaAs. It should be possible to grow films of ZB TcX directly on other ZB semiconductors substrates. The existence of large number of such ZB semiconductors and semiconducting substrates provides the opportunity to tune the lattice parameter and hence the band gap. Thus it is of interest to study the electronic and magnetic properties of these compounds. We have carried out a search of TcX compounds in order to ascertain the possibility of half-metallicity in these systems. It is hoped that the practical issues related to growth and physical property measurements of these compounds will be settled in near future. Successful fabrication of these materials offers exciting possibilities for the spintronic industry.

In this paper, we report systematic FP-LAPW calculations of binary ZB transition-metal compounds TcX (X=C, Si, Ge). We find HM ferromagnetism in these binary compounds at their optimized (equilibrium) cell volumes. We have thus investigated the electronic and magnetic properties for lattice parameters at and around these optimum (equilibrium) values. The remainder of this paper is organized as follows: In Section 2 we present the details of our calculations; in Section 3 we investigate the electronic and magnetic properties of these zinc blende binary Tc compounds TcX, where

X stands for C, Si and Ge. Exchange interactions and the Curie temperatures of these compounds are discussed in Section 4. Section 5 presents a summary of our results and conclusions.

## 2. Computational details

The ZB structure has the space group ( $F\bar{4}3m$ , or  $T_d^2$ , No. 216). The unit cell of TcX (X=C, Si and Ge) ZB structure may be viewed as an fcc lattice with four atoms per unit cell: Tc located at (0, 0, 0), X atoms at (0.25, 0.25, 0.25) and the two vacant sites at (0.5, 0.5, 0.5) and (0.75, 0.75, 0.75). We use the type-I AFM ordering [33] for the AFM calculations.

We used the WIEN2k [34] code, based on the full potential linear augmented plane wave plus local orbitals method and the density functional theory (DFT) [35]. The generalized gradient approximation (GGA) proposed by Perdew, Burke, and Ernzerhof (GGA-PBE96) was used for exchange and correlation [36] potentials. We consider full relativistic effects for the core states, and use the scalar approximation for the valence states. We have neglected the spin-orbit coupling because of its marginally small effect on the main results. We use 3000k points for the full Brillouin-zone integrations, using  $14 \times 14 \times 14$  mesh of *k*-points in the irreducible wedge of Brillouin zone according to the Monkhorst–Pack scheme [37]. Convergence was checked by increasing the number of *k* points. We set  $R_{mt} * K_{max}$  to 8.0 and considered angular expansion up to  $l_{max} = 10$  in the muffin tins. The self-consistent calculation was allowed to stop only after the integrated charge difference per formula unit,  $\int |\rho_n - \rho_{n-1}| dr$ , for the input and output charge densities  $\rho_{n-1}$  and  $\rho_n$  became less than  $10^{-4}$ .

## 3. Electronic structure of the binary compounds

We have carried out structural optimization of ZB TcX. For each of the binary compounds, we calculate total energy as a function of cell volume and thereby determine the equilibrium volume and lattice constant. Electronic densities of states (DOS), bands, and moments are then calculated for the equilibrium lattice constants. The equilibrium lattice constants *a*, moments per magnetic atom, the gaps  $G_{MAJ}$  of majority-spin bands, and the HM gaps  $G_{HM}$  of these binary compounds, are summarized in Table 1. Of all the binary Tc compounds based on IV we find three HMFs. Table 1 shows the energy gaps of the majority-spin bands. The total magnetic moment is mainly contributed by the Tc atoms, being the same as in ZB binary transition-metal compounds. A certain amount of magnetic moments is found in the interstitial region too. Unfortunately, no experimental or theoretical data of elastic constants are available for comparison.

Fig. 1 shows the spin-dependent total DOS of FM ZB TcX (X=C, Si and Ge) at their equilibrium lattice constants. We remark that all three compounds exhibit energy gap. X-*p* states occupy the part from  $-3.5$  to  $-1.5$  eV below the Fermi level, while X-*s* states

**Table 1**

The calculated equilibrium lattice constants *a* (Å), bulk modulus *B* (GPa), first order pressure derivative of bulk modulus *B'*, energy difference (meV)  $\Delta E_1 = E_{FM} - E_{FM}$  (between NM and FM states) and  $\Delta E_2 = E_{AF} - E_{FM}$  (between AFM and FM states), majority-spin gaps  $G_{MAJ}$  (eV), and HM gaps  $G_{HM}$  (eV) for the TcX compounds. The results are done with the predicted equilibrium lattice constant, respectively.

| Compounds | <i>a</i> | <i>B</i> | <i>B'</i> | $\Delta E_1$ | $\Delta E_2$ | $G_{MAJ}$ | $G_{HM}$ |
|-----------|----------|----------|-----------|--------------|--------------|-----------|----------|
| TcC       | 4.630    | 247.284  | 4.486     | 45.18        | 46.67        | 0.75      | 0        |
| TcSi      | 5.450    | 110.417  | 3.848     | 96.68        | 83.03        | 0.49      | 0.24     |
| TcGe      | 5.613    | 95.560   | 4.499     | 138.14       | 82.27        | 0.38      | 0.02     |

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