

First-principles theoretical studies of half-metallic ferromagnetism in CrTe

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Using full-potential linear augmented plane-wave method (FP-LAPW) we have studied the stability and electronic properties of the chalcogenide CrTe in three competing structures: rocksalt (RS), zinc blende (ZB), and the NiAs-type (NA) hexagonal. Although the ground state is of NA structure, RS and ZB are interesting in that these fcc-based structures, which can perhaps be grown on various semiconductor substrates, exhibit half-metallic (HM) phases above some critical values of the lattice parameter. We find that the NA structure is not HM at its equilibrium volume while both ZB and RS structures are. The RS structure is more stable than the ZB with an energy that is lower by 0.25 eV/atom. While confirming previous results on the HM phase in ZB structure, we provide hitherto unreported results on the HM RS phase, with a gap in the minority channel and a magnetic moment of 4.0 μ_B /f.u. A comparison of total energies for the ferromagnetic (FM), nonmagnetic, and antiferromagnetic (AFM) configurations shows the lowest energy configuration to be FM for CrTe in all the three structures. The exchange interactions in the RS and ZB structures are studied for a wide range of the lattice parameter using the linear-response method and a mapping of the total energy to the classical Heisenberg model. These linear-response calculations are performed in the linear muffin-tin orbitals (LMTOs) basis, using the atomic sphere approximation (ASA). We have verified that the results of the electronic structure obtained via the LMTO-ASA method under local-density approximation (LDA) and LDA+ U schemes are in close agreement with those obtained via the more accurate FP-LAPW method. The results show that the exchange interactions in the RS structure are much more short ranged than in the ZB structure. Hence, for the RS structure the exchange interactions are also studied by using a nearest- and next-nearest-neighbor (J_1 - J_2) model and the energy differences between FM and two AFM states. These J_1 - J_2 model results are obtained by using both the FP-LAPW and LMTO-ASA methods and compared with the linear-response results. The calculated Curie temperatures for the RS phase are consistently higher than those for the ZB phase.

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I. INTRODUCTION

Half-metallic (HM) ferromagnetism^{1,2} is an ideal property of materials from the viewpoint of their application in spintronic devices.³ Thus a considerable research has been devoted in recent years to the designing and measurement of the physical properties of such materials. In particular, some zinc-blende (ZB) structure transition-metal chalcogenides and pnictides have been predicted to be HM ferromagnets on the basis of theoretical calculations.^{4–11} Several Cr-based chalcogenides and pnictides,^{8,12–19} have been predicted or found to be not only HM but also exhibit high Curie temperatures T_c , sometimes in excess of 400 K. Sreenivasan *et al.*¹⁸ and Bi *et al.*¹⁹ have reported growing ~ 5 -nm-thick films of ZB CrTe using molecular-beam epitaxy on GaAs (100) substrate via a ZnTe buffer layer. Their measurements of temperature-dependent remanent-magnetization indicate a Curie temperature of ~ 100 K in these films. Two of the present authors have recently presented an extensive theoretical study of the exchange interactions and Curie temperatures in a series of Cr-based pnictides and chalcogenides in ZB structure as a function of the lattice parameter.²⁰ This study shows a robust HM ferromagnetism in ZB CrAs and

CrSb with high T_c over a wide range of the lattice parameter.

One important issue is that although the ZB phase of these compounds shows half metallicity and often (the promise of) high Curie temperature, the ground state usually has a different structure. For many of these compounds the ground state is NiAs-type hexagonal (NA). The semiconductor substrates on which thin films of these compounds are grown have cubic structures, usually rocksalt (RS) or ZB. The other point is that often the equilibrium volume does not show half metallicity, which appears only at an expanded volume. From this standpoint it would be useful to explore the possibility of the HM phase in structures other than ZB and, in particular, the RS structure. In this work we have examined this problem for CrTe. One attractive feature of bulk CrTe, at least from a theoretical viewpoint, is that the equilibrium volume in the ZB structure is already HM, whereas most other Cr-based chalcogenides and pnictides show half metallicity only at expanded volumes.⁸ Besides, the HM gap is larger in the chalcogenides such as CrTe or CrSe than in the pnictides such as CrAs or CrSb. This is due to larger Cr moment in the HM phase for the chalcogenides ($\sim 4 \mu_B$) than for the pnictides ($\sim 3 \mu_B$), which results in larger exchange splitting. Our previous study²⁰ has revealed that chalcogenides such as

TABLE I. Results obtained via the WIEN2K code: equilibrium lattice constant a , Cr-Te bond length L_{CrTe} , cohesive energy E_c , and bulk modulus B for ZB, RS, and NA CrTe in the FM state. E_c is defined as negative of the total energy of the formula unit with respect to the sum of the energies of the free Cr and Te atoms. E_1 , E_2 , and E_m are as defined in the text. All results shown are obtained using GGA. In some cases the corresponding LDA values are shown in parentheses. Experimental values of the lattice parameters for ZB CrTe films grown on GaAs (100) substrate via a ZnTe buffer layer (Refs. 18 and 19) and of bulk NA phase of CrTe (Ref. 23) are shown in square brackets.

Structure	a (Å)	L_{CrTe} (Å)	B (GPa)	E_c (eV)	E_1 (eV)	E_2 (eV)	E_m (eV)
ZB	6.263 (6.088) [6.1–6.21]	2.712	45.278 (57.962)	6.13	4.416	0.284	0.328
RS	5.727 (5.533)	2.864	59.744 (70.823)	6.38	1.605	0.019	0.076
NA ($c/a=1.542$) [1.557]	4.118 [3.997]	2.859	46.487	6.46	1.453	0.107	0

CrS and CrSe have a high likelihood of exhibiting antiferromagnetic (AFM) or complex magnetic behavior for a range of the lattice parameter, whereas such tendencies are less pronounced in CrTe. Hence, the focus of this work is on CrTe. We study the appearance of half metallicity in three competing structures for CrTe: NA, RS, and ZB. The ground-state structure (NA) shows half metallicity not at the equilibrium volume but at some expanded volume while both RS and ZB structures show half metallicity at their respective equilibrium (lowest energy) volumes. The RS structure is found to be more stable than the ZB with an energy that is lower by 0.25 eV/atom. Some band calculations for CrTe in the NA structure have already appeared²¹ in the literature and the same goes for ZB CrTe.^{6,8} Ahmadian *et al.*²² have recently discussed half metallicity at a ZB CrTe(001) surface and its interface with ZnTe(001). So far CrTe in the RS structure has not been studied from the viewpoint of its electronic and magnetic properties.

II. ELECTRONIC STRUCTURE

The two fcc-based structures ZB ($F\bar{4}3m$, No. 216) and RS ($Fm\bar{3}m$, No. 225) are well known. The NA structure ($P6_3/mmc$, No. 194) is based on a distorted hcp array of the arsenide anions. By contrast with the wurtzite structure, which shares the same formula type (AB) with the NA structure, the cations occupy all the octahedral sites rather than half the tetrahedral holes. There is one octahedral hole for each hcp lattice site and so the AB stoichiometry is preserved. The local coordinations of the anions and cations are different in the NA and wurtzite structures. Bulk samples of CrTe are known to crystallize in this structure²³ with cell parameters $a=3.997$ Å, $c=6.222$ Å at room temperature. The Cr-Te bond length is 2.78 Å and the c/a ratio is 1.557, different from the ideal hcp value of 1.633. Thus the six Te atoms form a trigonally distorted octahedron around Cr. The Cr-Cr nearest-neighbor distance in the ab plane is $a=3.997$ Å while the out-of-plane distance is $c/2=3.111$ Å. Readers may consult Fig. 1 of Ref. 21 to view the structure.

For the electronic-structure study we have used the WIEN2K code,^{24,25} which employs the full-potential linear augmented plane waves (FP-LAPWs) plus local orbitals method. The generalized gradient approximation (GGA) pro-

posed by Perdew *et al.*,²⁶ as well as the usual local-density approximation (LDA), was used for exchange and correlation potentials. We consider full relativistic effects for the core states and use the scalar approximation for the valence states. Spin-orbit (S-O) coupling is neglected in obtaining the results reported in this paper, as it should have negligible effect on the results for CrTe.²⁷ About 3000 K points were used for the Brillouin-zone (BZ) integrations, using the Monkhorst-Pack scheme²⁸ and $14 \times 14 \times 14$ divisions of the BZ. We used $R_{mt} \times K_{max}=8.0$ and angular momentum expansion up to $l_{max}=10$ in the muffin tins and $G_{max}=14$ for the charge density. For the calculations involving the different structures the muffin-tin (MT) radii were taken to be equal to 2.2 and 2.5 bohr for Cr and Te atoms, respectively. The same MT radii were used in the calculations of different structures at their equilibrium volumes in order to compare accurately the total energies. Self-consistency in charge density was achieved to an accuracy higher than 10^{-4} .

We carried out a systematic structural optimization of NA, ZB, and RS CrTe in ferromagnetic (FM), nonmagnetic (NM) (nonspin polarized), and AFM configurations. For the NA structure the c/a ratio was optimized and the result $c/a=1.542$ for the FM phase is in good agreement with previously obtained result.²² This optimized c/a ratio was used for the final NA structure volume optimization. Bulk modulus was calculated by fitting the volume vs energy results to the empirical Murnaghan equation of state.²⁹ We find that for all three structures the FM phase has lower energy than the NM and AFM phases. To assess the relative stability of CrTe in different structures we consider the total energy per formula unit (f.u.) in the ZB and RS structures relative to the NA structure. The calculated optimized equilibrium lattice constant a , bulk modulus B , the total energy difference E_1 between the NM and the FM states: $E_1=E_{\text{NM}}-E_{\text{FM}}$, and E_2 between the AFM and the FM states: $E_2=E_{\text{AFM}}-E_{\text{FM}}$, and the metastability energy E_m , which is the total energy per formula unit relative to the NA structure, are given in Table I. Note that for RS and ZB structures, in calculating E_2 we have considered the energies of three AFM configurations: [001], [110], and [111] and considered the lowest of these energies. For the hexagonal NA structure, the AFM state was created by simply ascribing equal but opposite magnetic moments to the two Cr atoms in the unit cell. Cohesive properties are dependent on the treatment of the exchange-

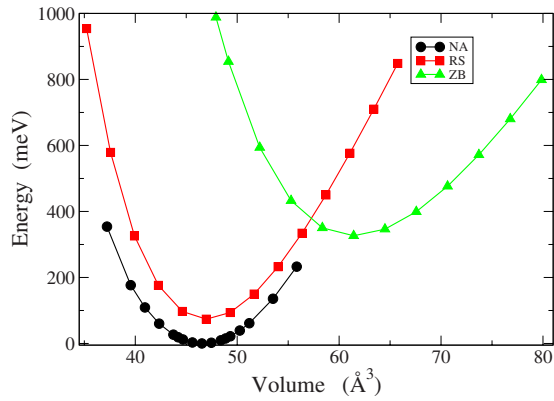


FIG. 1. (Color online) Energy as a function of volume per atom in the three-structural phases of CrTe, obtained via FP-LAPW GGA WIEN2K method.

correlation effects in a nontrivial manner, as indicated by the LDA values of the lattice parameter and bulk modulus shown in parentheses in Table I for the RS and ZB equilibrium structures, where all other values are from GGA. Increased correlation effects in GGA increases the equilibrium lattice parameter and lowers the bulk modulus.

It is apparent from Table I that the FM state is lower in energy than the NM and AFM states for all the three structures and the ground-state structure is NA. Our calculated CrTe results of ZB and NA by GGA are in good agreement with previous results.^{6,22} The calculated total energy as a function of the atomic volume for FM phase ZB, RS, and NA structures of CrTe is shown in Fig. 1. The ZB and RS phases are higher in energies than NA by 0.328 eV and 0.076 eV/atom, respectively. However, the RS structure is only marginally unstable against the NA structure, and more stable than the ZB structure, with an energy that is about 0.25 eV/atom lower. Thus we have carried out a detailed study of the RS phase. According to our calculation the ZB phase is more stable than the RS phase for volumes per atom greater than 57 Å³ (Fig. 1). It should be possible to stabilize the RS phase at lower volumes. Recently Sreenivasan *et al.*¹⁸ and Bi *et al.*¹⁹ were able to grow ZB CrTe films on GaAs substrates. Their measured values of the lattice parameter lie in the range 6.1–6.21 Å, corresponding to volumes per atom 57–59.9 Å³. Our calculations suggest that in order to grow RS CrTe one should consider suitable substrates with lattice parameter less than 6.1 or 6.0 Å.

As the density of states (DOS) for the ZB structure has been described in several publications,^{6,8} we present only the DOSs for the RS and NA structures in this work. For the RS structure the majority-spin channel, as in case of the ZB structure, is metallic while the minority-spin states have an energy gap of width 0.76 eV around the Fermi level. The minority-spin gap for the ZB structure is 2.9 eV. All of these values refer to the respective equilibrium volumes. Thus both ZB and RS phases are HM systems. The HM gap, which is defined in Ref. 6 is 0.83 eV for the equilibrium ZB volume, while for the RS equilibrium volume it is very small, being about 0.03 eV. However, it increases with increasing volume. The spin polarization at the Fermi level is quite small for RS CrTe but the half metallicity is robust against volume expan-

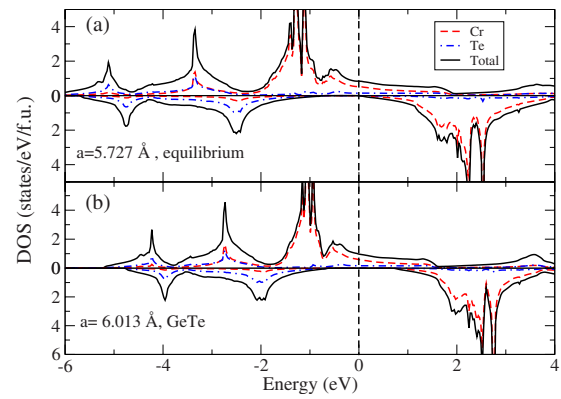


FIG. 2. (Color online) DOS for the RS structure, obtained via the FP-LAPW GGA method, at (a) the equilibrium lattice parameter $a=5.727$ Å for CrTe, (b) expanded lattice parameter $a=6.013$ Å, the equilibrium value for RS-GeTe, a suitable substrate for growing RS CrTe.

sion. The general structure of DOS is similar around the Fermi level for both structures. A closer inspection, however, reveals some differences. In the spin-up channel there is a gap in the energy interval between 1.62 and 2.82 eV just below the Fermi level for the ZB structure. On the other hand, for the spin-down DOS the gap at the Fermi level in ZB CrTe is larger than that in RS CrTe. Also the peak structures differ by an energy shift with respect to the Fermi level. All these differences should be accountable on the basis of different levels of hybridization between the Cr 3d and Te 5p orbitals and different exchange splittings for the two structures. The growth of metastable ZB-CrAs,¹⁴ ZB-CrSb,¹⁷ and ZB-CrTe (Refs. 18 and 19) indicates the suitability of RS-CrTe for growth on carefully chosen substrates. The equilibrium lattice constant of RS CrTe (5.727 Å) is only 4.7% smaller than the equilibrium lattice parameter 6.013 Å of RS GeTe (both values quoted are from calculations). Thus RS GeTe should act as a suitable substrate for growing RS CrTe films.

In Figs. 2 and 3 we show the DOSs for the RS and NA structure CrTe at their respective equilibrium volumes ob-

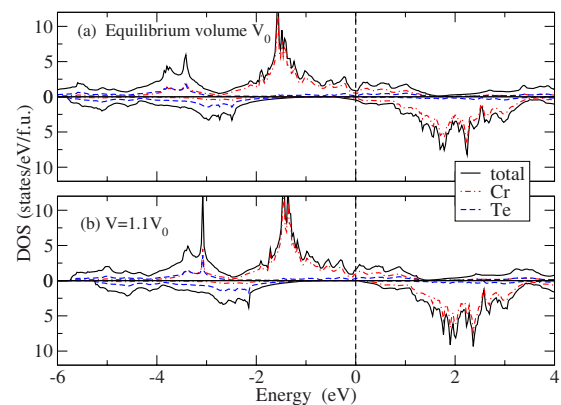


FIG. 3. (Color online) DOS for the NiAs structure at the (a) equilibrium volume V_0 ($c/a=1.5424$, $a=4.118$ Å) and (b) 10% expanded volume $V=1.1V_0$ ($c/a=1.5424$), obtained via the FP-LAPW GGA method.

TABLE II. Comparison of magnetic moments per formula unit (μ_B), minority-spin gap G_{\min} (eV), and HM gap G_{HM} (eV) for FM CrTe in ZB, RS, and NA structures, at their respective equilibrium volumes, obtained by using the FP-LAPW GGA method WIEN2K. The numbers without and within the parentheses refer to values obtained by using GGA and LDA, respectively.

Structure	Cr	Te	Interstitial	Total	G_{\min}	G_{HM}
ZB	3.542 (3.501)	-0.157 (-0.119)	0.616 (0.619)	4.000 (4.000)	2.911	0.829
RS	3.563 (3.517)	-0.112 (-0.079)	0.548 (0.548)	4.000 (3.986)	0.761	0.028
NA	3.489 (3.398)	-0.105 (-0.076)	0.520 (0.503)	3.904 (3.825)		

tained via WIEN2K FP-LAPW code. Note that at equilibrium volume CrTe in NA structure is not HM, but becomes so at expanded volumes, provided the c/a ratio stays unchanged. In Fig. 2(b), the DOS of RS CrTe at the equilibrium RS-GeTe lattice parameter (6.013 Å) is given, where it is seen to be HM. The minority-spin gap is 1.388 eV, HM gap is 0.637 eV. Both the minority-spin gap and HM gap become larger with increasing lattice parameter and half metallicity is robust with respect to lattice expansion.

The calculation of the exchange interactions to be presented in Sec. IV are based on the Green's function and multiple-scattering formalism and implemented in the tight-binding (TB) linear muffin-tin orbitals-atomic sphere approximation (LMTO-ASA) basis.³⁰⁻³² In order to ensure that the LMTO results are in close agreement with those given by the FP-LAPW method we repeated the electronic DOS and magnetic-moment calculations using the TB-LMTO-ASA method. The exchange-correlation potential of Vosko *et al.*³³ was used for these calculations. We optimize the ASA errors by including empty spheres in the unit cell.

Results of the electronic structure are strongly dependent on the treatment of the exchange-correlation effects. Enhanced correlation generally increases the equilibrium lattice parameter and the magnetic moment for a given volume. The equilibrium lattice parameter for the ZB structure is 6.26 Å according to the FP-LAPW calculations using GGA, 6.09 Å according to FP-LAPW and LDA, and 6.03 Å according LMTO-ASA method using LDA. These LDA values value are in close agreement with the result 6.07 Å from the Korringa-Kohn-Rostoker (KKR) Green's-function method and LDA.⁸ For RS CrTe equilibrium lattice parameter from FP-LAPW GGA is 5.727 Å, while replacing GGA by LDA results in the value 5.53 Å, a decrease in about 3%. Since GGA is not incorporated at this stage in the TB-LMTO-ASA code that we used, we checked the effect of enhancing the correlation effect in LMTO-ASA by using the LDA+ U method.³⁴ The value of U to be used for Cr 3d orbital is difficult to know *a priori*. So we have used it as an adjustable parameter. The width of the energy gap in the minority states for the RS structure has a noticeable dependence on the treatment of exchange and correlation effects. This gap is widened with enhancement of correlation effects. For a lattice parameter just above the critical value for the HM state the LDA calculation gives a gap width of approximately 0.5 eV, while for the LDA+ U calculation with $U=0.075$ Ry (1.02 eV), the gap increases to ~ 0.8 eV, which compares well with the FP-LAPW GGA value of 0.76 eV. Hence, this value of $U=0.075$ Ry is used as a token to examine the effect of

enhanced correlations on the values of the exchange interactions and Curie temperature T_c calculated via the LMTO-Green's-function method (to be discussed in Sec. IV). The equilibrium lattice parameter for RS CrTe increases from 5.245 to 5.4 Å, i.e., by about 3%, from LDA to LDA+ U with $U=0.075$ Ry (and $J=0$, see Ref. 34 for details). In general, the electronic-structure results obtained via the TB-LMTO-ASA method are in close agreement with those given by the FP-LAPW method.

III. MAGNETIC MOMENTS

Results of the WIEN2K calculations for magnetic moments and some other useful quantities are given in Table II. The calculated total magnetic moment for both ZB and RS structures are 4.000 μ_B /f.u. at their equilibrium lattice parameters, indicating complete half metallicity. For NA CrTe half metallicity appears at a volume higher than the equilibrium volume [Table II and Fig. 3(b)]. The moment versus volume variations for the three structures obtained in the WIEN2K calculations are shown in Fig. 4. The magnitudes of the moments on the two sublattices increase with increasing lattice parameters, due to decreased hybridization between Cr d and Te sp orbitals. Above a critical value of the lattice parameter, the moment per f.u. saturates at a value of 4.000 μ_B , as the HM state is achieved, while the local moments on the Cr and Te sublattices increase in magnitude, remaining opposite to each other. Note that the minority-spin gap G_{\min} in Table II appears drastically different for the RS and ZB CrTe for two reasons: (i) for a given lattice parameter the Cr-Te bond length is shorter in ZB than in RS CrTe, resulting in in-

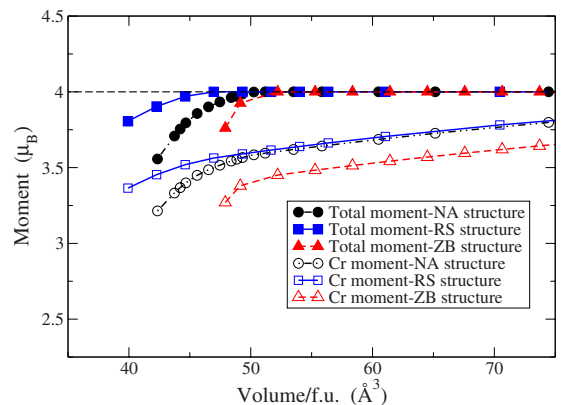


FIG. 4. (Color online) Magnetic moment per f.u. for the NA, ZB, and RS structures obtained via FP-LAPW GGA calculations.

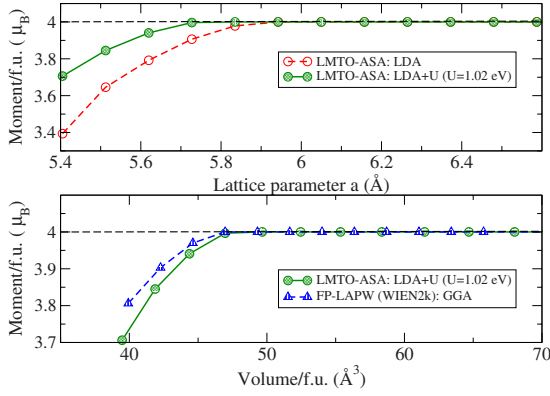


FIG. 5. (Color online) Magnetic moment per f.u. versus lattice parameter for RS CrTe calculated by using LMTO-ASA method and LDA and LDA+ U ($U=1.02$ eV, $J=0$) schemes (upper panel). Enhanced correlation effects incorporated via the LDA+ U method brings the results in close agreement with those obtained via FP-LAPW-GGA method (lower panel).

creased Cr-Te hybridization for the ZB case and (ii) the values quoted in Table II are for the respective equilibrium lattice parameters: 5.727 Å for RS and 6.263 Å for ZB CrTe. The value of G_{\min} for RS CrTe calculated at $a=6.263$ Å, the equilibrium ZB lattice parameter, would be 1.8 eV.

In LMTO-ASA method the interstitial region is described by empty spheres and the electronic properties of this region is primarily dictated by the tails of the orbitals centered at the real atoms such as Cr and Te. As in the FP-LAPW calculation, we find that the magnetic moments of the empty spheres are parallel to those of the Cr atoms, while those of the Te atoms are antiparallel. For the RS structure, the moment on the Te atom is small (around 1–2 % of that of the Cr atom) for low values of the lattice parameter, while the moment on the two empty spheres are each about 3.5–4 % of that of the Cr atom. With increasing lattice parameter, the Te moment increases with respect to the Cr moment to about 7–8 % at the highest lattice parameter studied. At these high lattice parameters the moments on the empty spheres, originating from the tails of the nearby Cr $3d$ orbitals, drop to about 2% of the Cr moment. In the ZB structure, the two empty spheres are not equivalent and the induced moments on the empty spheres and the Te atoms as well as the variation in the total and Cr moments with the lattice parameter in this case have been described in detail in our recent publication.²⁰

Magnetism is strongly dependent on correlation effects. Even in the metallic states, where magnetism is believed to be described reasonably well by density-functional methods and LDA, different treatment of the exchange-correlation effects lead to different values of magnetic moments. We thus compare the magnetic moments of our LMTO-ASA calculations for LDA and LDA+ U .³⁴ The enhanced correlation effects in the LDA+ U calculations (using $U=0.075$ Ry, $J=0$) increase the values magnetic moments and bring the results in general close agreement with those obtained via using GGA in the WIEN2K package. This is shown in Fig. 5. The agreement between GGA and LDA+ U results is due to the fact that the GGA goes beyond the LDA by including electron correlations in some way.

IV. EXCHANGE INTERACTION AND CURIE TEMPERATURE

We calculate the exchange interactions by mapping^{35,36} the system energy onto a classical Heisenberg model

$$H_{\text{eff}} = - \sum_{ij} J_{ij} \mathbf{e}_i \cdot \mathbf{e}_j, \quad (1)$$

where i, j are site indices, \mathbf{e}_i is the unit vector pointing along the direction of the local magnetic moment at site i , and J_{ij} is the exchange interaction between the moments at sites i and j . In this work, we use the method of Liechtenstein *et al.*,³⁷ based on multiple-scattering formalism, which was later implemented for random magnetic systems by Turek *et al.*,³⁸ using coherent-potential approximation (CPA) and the TB-LMTO method. The Curie temperature T_c is calculated in both the mean-field approximation (MFA) and the random-phase approximation (RPA) (Ref. 39) from the exchange interactions and their lattice Fourier transform. For a discussion of the various methods used in the mapping procedure the reader may consult our previous publication.²⁰ T_c^{RPA} can be shown to be smaller than T_c^{MFA} ,³⁶ and has been found to be in close agreement with results given by Monte Carlo simulations.⁴⁰ Unless otherwise stated, the T_c estimates given in this paper are based on RPA.

Sandratskii *et al.*³⁵ have discussed the case when, in addition to the interaction between the strong moments, there is one secondary, but much weaker, interaction between the strong and one induced moment. In this case, the Curie temperature, calculated under the MFA, is enhanced due to this secondary interaction, irrespective of the sign of the secondary interaction. In other words, the Curie temperature would be somewhat higher than that calculated by considering only the interaction between the strong moments. The corresponding results under the RPA have to be obtained by solving two equations simultaneously. One can assume that the RPA results for the Curie temperature follow the trends represented by the MFA results, being only somewhat smaller, as observed in the absence of induced moments.

In Fig. 6 we compare the exchange interactions between the Cr atoms obtained for the equilibrium lattice parameter of 5.727 Å (FP-LAPW GGA result) of RS CrTe with those in ZB CrTe for two values of the lattice parameter, one above and the other below the value 5.727 Å. For the RS structure the interactions are all FM while for the ZB structure they are predominantly AFM in this range of the lattice parameter. In our previous publication we have shown that due to these AFM interactions, the Curie temperature in ZB CrTe is drastically reduced for a certain range of the lattice parameter. The ground state for this structure may even have a more complex magnetic phase than simple FM. Thus for lattice parameters around 5.74–5.75 Å RS CrTe is not only guaranteed to be a HM ferromagnet but also should have a higher T_c than ZB CrTe of similar volume per atom.

In Fig. 7 we compare the T_c^{RPA} values for RS and ZB CrTe obtained by using the TB-LMTO-Green's-function method and using only the exchange interactions between the Cr atoms (strong moments). The reference state used for the calculation is the FM state. For the ZB phase, the use of Eq.

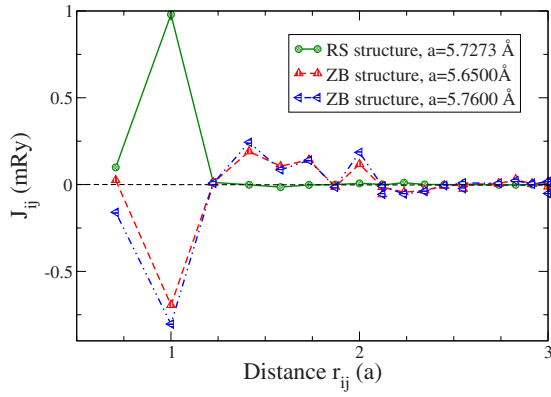


FIG. 6. (Color online) Exchange interaction, computed by using the LMTO-Green's-function method between the Cr atoms in the ZB and RS structures as a function of the lattice parameter. The two lattice parameters for the ZB structure chosen are above and below the equilibrium lattice parameter 5.727 Å for the RS structure, obtained in the FP-LAPW GGA (WIEN2K) calculation.

(4) of Ref. 20 neglecting all the induced moments, results in unphysical negative T_c in the lattice parameter range ~ 5.7 – 6.0 Å because of dominant AFM Cr-Cr exchange at the nearest- and next-nearest-neighbor sites. Since the exchange calculations are done with respect to a “FM” reference state with parallel magnetic moments, negative values of the exchange interactions, and, in particular, the exchange constant $J_0 = \sum_j J_{0j}$, imply instability of the reference FM state. The ZB phase has been discussed in detail in our previous publication,²⁰ where we showed that in spite of these nearest- and next-nearest-neighbor AFM interactions, the FM state is lower in energy than the AFM[001] and AFM[111] states. The cumulative effect of the interactions involving the

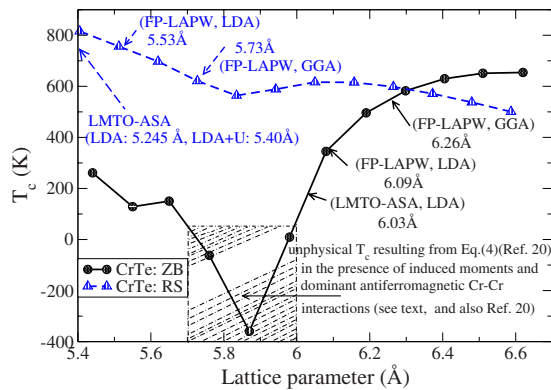


FIG. 7. (Color online) The RPA Curie temperature as a function of the lattice parameter for the ZB and RS structures of CrTe, calculated by considering the Cr-Cr interaction only and by using the FM reference state. The arrows indicate the equilibrium lattice-parameter values according to various calculations. The results for T_c are somewhat affected by the “induced” moments on the Te atoms as well as the empty spheres used in the LMTO Green's-function calculation. For ZB CrTe, this error, combined with strong AFM spin fluctuation for the lattice parameters in the range ~ 5.7 – 6.0 Å, result in unphysical negative values of T_c in this range (see text and also the discussion in Ref. 20 for details). Away from this range T_c values are reliable, although somewhat underestimated.

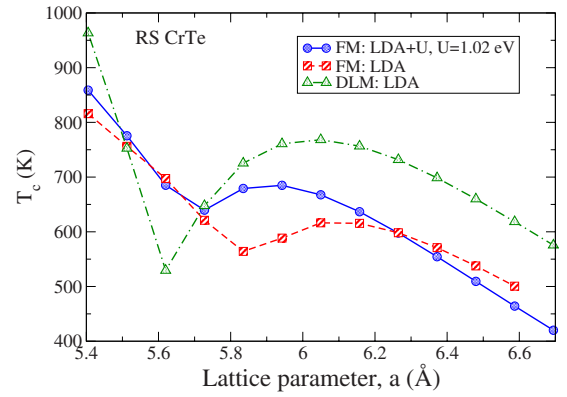


FIG. 8. (Color online) Comparison of the Curie temperatures in RS CrTe calculated using RPA and LMTO Green's-function method for FM reference states with LDA and LDA+U, and DLM reference states with LDA.

more distant pairs of Cr atoms, and particularly those due to the induced moments, may stabilize the FM phase. Alternatively, the magnetic state is possibly complex, different from FM, AFM[001], and AFM[111]. If the magnetic state is indeed FM, then according to the prescription of Sandratskii *et al.*³⁵ T_c should be enhanced with respect to the value given by Eq. (4) and may be positive but small. Note that Sreenivasan *et al.*¹⁸ and Bi *et al.*¹⁹ report the lattice parameters for their ZB CrTe thin films grown on GaAs substrates to be in the range 6.1–6.21 Å and their measurements of remanent magnetization indicate $T_c \sim 100$ K. This is not too far off from the calculated values 200–400 K shown in Fig. 7. For RS CrTe, on the other hand, all dominant interactions are FM, and as a result the T_c calculated by using Eq. (4) of Ref. 20 and based on only Cr-Cr interactions give positive values. The actual T_c 's are most probably higher than what is shown in Fig. 7. The effect of enhanced correlation for RS CrTe incorporated via LDA+U method, using $U=0.075$ Ry, and $J=0$, is shown in Fig. 8. Within the uncertainties of our results there seems to be no appreciable difference between the predictions of LDA and LDA+U. Note that all results for T_c are calculated by using interactions up to 111 shells of neighbors to ensure convergence of the calculated values.

In our previous publication²⁰ we have argued that estimates of T_c from above the (paramagnetic \leftrightarrow ferromagnetic) transition can be obtained by considering the reference state for the exchange calculation in the method of Liechtenstein *et al.*³⁷ to be given by the disordered local moments (DLM) (Ref. 41) model. In a collinear magnetic model, this disordered local-moment state describes the paramagnetic state. In this description magnetic moments appear only on the magnetic, i.e., Cr, atoms, but are oppositely directed for half of the randomly chosen atoms, giving a zero moment in total. The randomness is treated using the CPA.^{31,32} T_c given by the DLM reference states are usually higher than their FM counterparts. The DLM estimates of T_c are free from errors originating from the neglect of the exchange interactions involving induced moments in Eqs. (3) and (4) of Ref. 20. For this reason we have computed the T_c using the DLM model as well and compare these with the T_c obtained for the FM

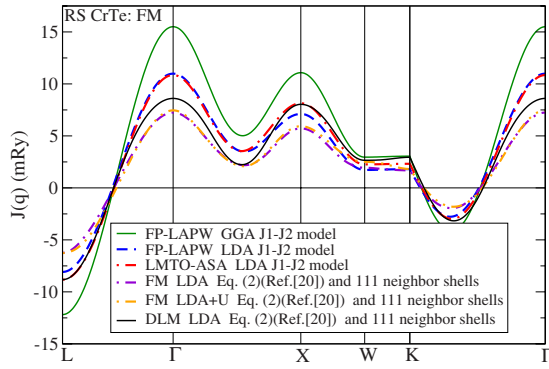


FIG. 9. (Color online) Comparison of the lattice Fourier transforms of the Cr-Cr exchange interactions in RS CrTe for the lattice parameter 5.727 \AA and the various cases discussed in the text.

reference states in Fig. 8. As expected, the DLM values are usually higher for most of the lattice parameters. Since the ground state of RS CrTe is known to be FM for the entire range of the lattice parameters studied, we expect the FM results to be valid, although somewhat underestimated due to the neglect of the induced moments. DLM estimates should decrease somewhat if electronic entropy effects are incorporated.⁴² Thus the theoretical estimates of T_c should lie between those given by the FM and DLM results.

The exchange interactions for RS and ZB CrTe shown in Fig. 6 were obtained using the multiple-scattering method of Liechtenstein *et al.*³⁷ For the RS structure the nearest- and next-nearest-neighbor exchange interactions (J_1 and J_2) appear to dominate all other distant interactions while for the ZB structure the interactions definitely go beyond the first two neighbors. It would thus transpire that a J_1 - J_2 model may suffice to describe the magnetism of the RS phase, whereas such a model would clearly be inadequate for the ZB phase. As the calculations of exchange interactions are often based on the energy differences between the FM and a few chosen AFM configurations and numerous such calculations have appeared in the literature, we decided to use the RS CrTe as a test case to check the validity of such calculations. For the RS phase, if one is prepared to neglect interactions beyond the next-nearest-neighbor pairs, then the nearest-neighbor interaction J_1 and the next-nearest interaction J_2 can be obtained from the energies of the FM and AFM[001] and AFM[111] states.⁴³ We have calculated J_1 and J_2 using this procedure for the FP-LAPW-GGA equilibrium lattice parameter of 5.727 \AA and the results are: $J_1=3.76 \text{ meV}$, $J_2=27.63 \text{ meV}$ (FP-LAPW-GGA), $J_1=3.29 \text{ meV}$, $J_2=18.32 \text{ meV}$ (FP-LAPW-LDA), and $J_1=4.61 \text{ meV}$, $J_2=20.0 \text{ meV}$ (LMTO-ASA-LDA). One could argue that all the distant-neighbor interactions between the Cr atoms given by the multiple-scattering method get mapped effectively on to just two interactions in the J_1 - J_2 model. This is similar to trying to reproduce the band structure of a long-ranged Hamiltonian via a TB model with only nearest- and next-nearest-neighbor interactions. A meaningful way to compare the two approaches may be to compute the dispersion $J(\mathbf{q})=\sum_{\mathbf{R}} J_{0\mathbf{R}}^{\text{Cr,Cr}} \exp(i\mathbf{q}\cdot\mathbf{R})$. In Fig. 9 we show these dispersion curves for RS CrTe along the various symmetry directions and all the different cases considered. The

values of the exchange interactions quoted above for the J_1 - J_2 model would put the T_c for RS CrTe with lattice parameter 5.727 \AA around $\sim 1000 \text{ K}$. A possible reason that the magnitudes of the $J(\mathbf{q})$ values based on the multiple-scattering formalism and FM reference state come out smaller than those given by the J_1 - J_2 model is that in computing the former results [based on Eq. (3) of Ref. 20] we have neglected all interactions involving the induced moments. A more plausible scenario is that calculations based on the energy differences between various magnetic configurations tend to (under) overestimate the exchange interactions and T_c . Such a conclusion seems to be also supported in a recent study by Fischer *et al.*⁴³ In particular, calculations where only a few magnetic configurations (based on the assumption of short-ranged exchange) are used are of doubtful quantitative validity. Note that even though the exchange interactions in Fig. 6 appear to be negligible beyond the distance of $\sim 2a$, their cumulative effect on the calculated T_c is non-negligible. The results for T_c shown in Figs. 7 and 8 were all calculated by considering interactions up to 111 shells of neighbors to ensure convergence.

V. SUMMARY OF RESULTS

We have provided a comparison of the magnetic properties of CrTe in three different structures: NA, RS, and ZB, at their respective equilibrium volumes. A detailed comparison of the HM properties of the ZB and RS phases, as a function of the lattice parameter, is provided, as thin films grown on most substrates are likely to have either of these two structures. The RS phase is more stable than the ZB phase, in that the lowest energy state in the RS phase is about 0.25 eV/atom lower than that for the ZB phase. For some range of the lattice parameter, namely, between 5.6 – 6.1 \AA , there are significant AFM interactions between the Cr atoms in the ZB phase. The RS phase appears to be free from such strong AFM interactions and thus ferromagnetism appears to be more robust in the RS phase than in the ZB phase. Increased AFM interactions in the ZB phase result from the smaller distance between the Cr and the Te atoms and consequently increased hybridization between the Cr $3d$ and Te $5p$ orbitals in the ZB phase than in the RS phase for the same lattice parameter. This results in a lower Curie temperature T_c for the ZB structure over a wide range of the lattice parameter. Increased stability with respect to the ZB phase, stronger ferromagnetism, and the promise of a higher T_c should prompt the experimentalists to try to fabricate CrTe thin films in the RS structure. Our results suggest that it should be possible to stabilize RS CrTe films on suitable substrates that provide lattice parameters less than 6.1 – 6.0 \AA .

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