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Density functional study of electronic, charge density, and chemical bonding properties of 9-methyl-3-Thiophen-2-Yl-Thieno [3,2-e] [1, 2, 4] Thiazolo [4,3-c] pyrimidine-8-Carboxylic acid ethyl ester crystals

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ABSTRACT

A comprehensive theoretical density functional investigation of the electronic crystal structure, chemical bonding, and the electron charge densities of 9-Methyl-3-Thiophen-2-Yl-Thieno [3, 2-e] [1, 2, 4] Thiazolo [4,3-c] Pyrimidine-8-Carboxylic Acid Ethyl Ester ($C_{15}H_{12}N_4O_2S_2$) is performed. The density of states at Fermi level equal to 5.50 (3.45) states/Ry cell, and the calculated bare electronic specific heat coefficient is found to be 0.95 (0.59) mJ/mole-K² for the local density approximation (Engel-Vosko generalized gradient approximation). The electronic charge density space distribution contours in (1 0 0) and (1 1 0) planes were calculated. We find that there are two independent molecules (A and B) in the asymmetric unit exhibit intramolecular C–H...O, C–H...N interactions. This intramolecular interaction is different in molecules A and B, where A molecule show C–H...O interaction while B molecule exhibit C–H...N interaction. We should emphasis that there is π – π interaction between the pyrimidine rings of the two neighbors B molecules gives extra strengths and stabilizations to the superamolecular structure. The calculated distance between the two neighbors pyrimidine rings found to be 3.345 Å, in good agreement with the measured one (3.424(1) Å).

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1. Introduction

Generally compounds containing fused pyrimidine ring are a broad class of heterocycles that have received much attention in recent years due to their wide range of biological activities [1] such as antiviral [2], analgesic activities [3], antimicrobial [4], inhibition of cancer cell proliferation [5,6], antitumor [7], anti-inflammatory activities [8], antagonism of α_1 adrenoceptors [9], and prevention of cartilage destruction in articular diseases [10]. Antiviral and antimicrobial agents [2] are among the most commonly prescribed pharmaceuticals worldwide [2]. Recently Farshori et al. [11] and Bala et al. [12] found that pyrimidine derivatives are widely used as an antifungal agent for the treatment of candidiasis due to fact that pyrimidine derivatives possess anticandidal activity. In the last few

years and due to the hard research work of Vorobev et al. [13] and Shishoo et al. [14] pyrimidine and thieno-pyrimidine have become a well sought-privileged class of compounds in drug discovery programs. The enhancing of therapeutic significance of these agents has a consequence in identification of more potent compounds which exhibit several potentially useful and attractive biological activities [2,15,16]. These potent compounds show dihydrofolate reductase inhibition and antitumor [17,18], diuretic properties [19], antimicrobial [20], antiviral, and cytotoxic activities [21,22].

The centrosymmetrically 9-Methyl-3-Thiophen-2-Yl-Thieno [3,2-e] [1, 2, 4] Thiazolo [4, 3-c] Pyrimidine-8-Carboxylic Acid Ethyl Ester ($C_{15}H_{12}N_4O_2S_2$) crystals has been synthesized by Fathima et al. [1]. It was subjected to single crystal X-ray diffraction analysis so that its supra-molecular structure could be investigated in terms of possible intermolecular interactions. The supramolecular packing will be governed by steric interactions and van der Waals forces. Generally in the crystals structures, the molecules are linked by noncovalent intermolecular interactions such as electrostatic interactions, hydrogen bonds, and van

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der Waals interactions [23,24]. Molecules tend to undergo shape simplification during the process of crystal growth, which gives rise to dimers and then to high-order aggregates in order to adapt to a close-packing in the solid state. The high tendency of achiral molecules to crystallize centrosymmetrically could be due to such a close-packing driving force.

From above and due to the attractive properties and activities of pyrimidine and thieno-pyrimidine we have taken the motivation and put more attention to investigate the electronic structure, space charge density and chemical bonding properties of 9-Methyl-3-Thiophen-2-YI-Thieno [3,2-e] [1, 2, 4] Thiazolo [4,3-c] Pyrimidine-8-Carboxylic Acid Ethyl Ester crystals by means of density functional theory. The unit cell parameters and the atomic positions have been taken from X-ray diffraction of the newly synthesized crystal [1].

In the next section, we briefly describe the calculation procedure and give the computational details. In Section 3, the discussion of our results for structural properties was reported. Finally, conclusions will be given in Section 4.

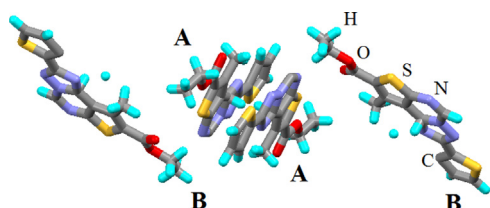


Fig. 1. The packing of molecules in the crystal structure of 9-Methyl-3-Thiophen-2-YI-Thieno [3,2-e] [1, 2, 4] Thiazolo [4,3-c] Pyrimidine-8-Carboxylic Acid Ethyl Ester crystals; it is illustrated there are two different molecules A and B.

2. Structural properties and computational details

We make use of the X-ray crystallographic data¹ which was taken from Cambridge Crystallographic Data Center (CCDC 787335). The packing of molecules of the centrosymmetrically 9-Methyl-3-Thiophen-2-YI-Thieno [3,2-e] [1, 2, 4] Thiazolo [4,3-c] Pyrimidine-8-Carboxylic Acid Ethyl Ester crystals structure is illustrated in Fig. 1. The crystal structure contains two independent molecules A and B in the asymmetric unit (Fig. 1). The C–H...N interaction forms centrosymmetric head to head dimers and the same for C–H...O interaction [1]. To solve the Kohn Sham DFT equations [25] we have performed comprehensive investigation using the all-electron full potential linearized augmented plane wave (FP-LAPW) method within the framework of the WIEN2K code [26]. Two different possible approximations for the exchange correlation (XC) potentials were employed. These are the local density approximation (LDA-CA) [27] and Engel–Vosko generalized gradient approximation (EV-GGA) [28], which are based on exchange-correlation energy optimization to calculate the total energy. The crystal structure of the title compounds were optimized by minimization of the forces (1 mRy/au) acting on the atoms using Perdew–Burke–Ernzerhof generalized gradient approximation (we call this GGA) [29]. The electronic structure, electron space charge density and the chemical bonding were calculated from the relaxed geometry. Muffin-tin radius of all atoms was chosen in such a way that the spheres did not overlap. To achieve the total energy convergence, the basis functions in the interstitial regions were expanded up to $R_{\text{mt}} \times K_{\text{max}} = 7.0$ and inside the atomic spheres for the wave function. The maximum value of l were taken as $l_{\text{max}} = 10$, while the charge density is Fourier expanded up to $G_{\text{max}} = 20$ (a.u)⁻¹. We have used 35 k-points in the irreducible Brillouin zone for structural optimization. For the calculation of electronic properties 180 k-points were used.

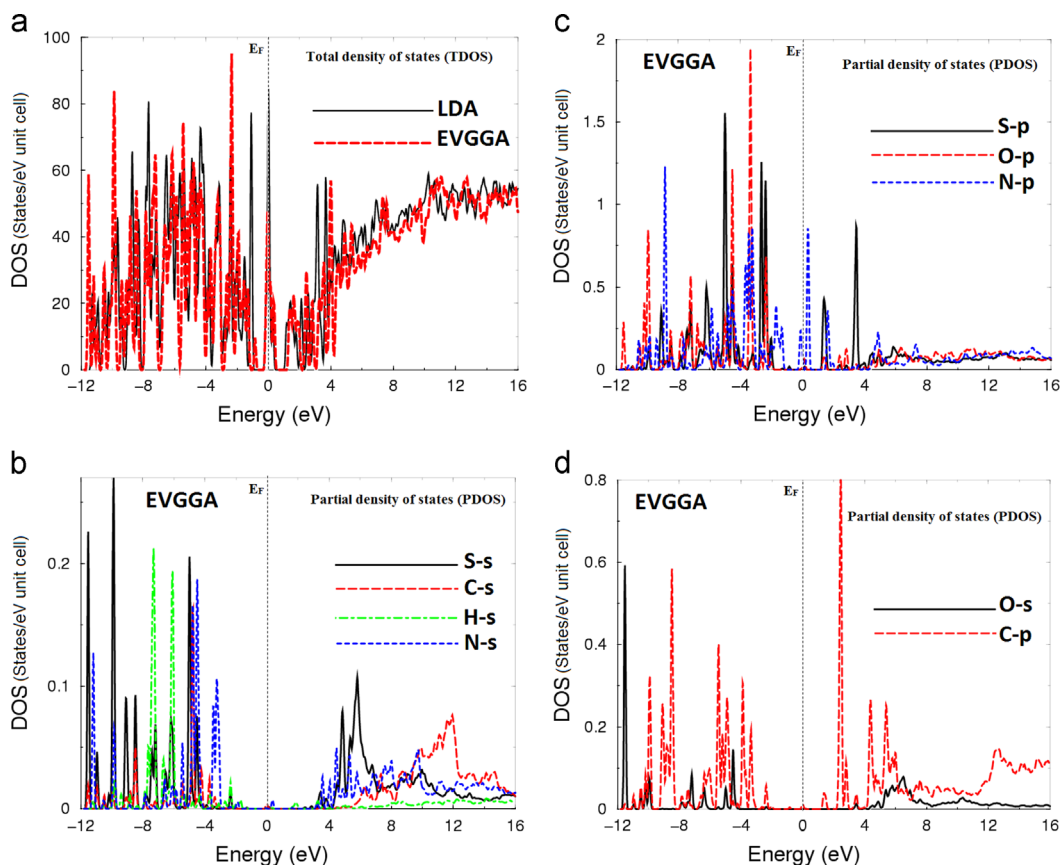


Fig. 2. Calculated total and partial densities of states (states/eV unit cell). The zero energy is arbitrarily taken at the Fermi level.

3. Results and discussion

3.1. Band structure and density of states

The angular momentum decomposition of the atoms projected density of states and the electronic band structure dispersion in

k-space along the high symmetry directions in the irreducible Brillouin zone were presented in Figs. 2 and 3, where the zero energy is taken at the Fermi level. The calculations were done using LDA and EVGGA. Using EVGGA the Fermi energy (E_F) is situated at 0.16636 eV while with LDA it is situated at 0.14192 eV. It is clear the EVGGA causes to shift the E_F towards lower energy by

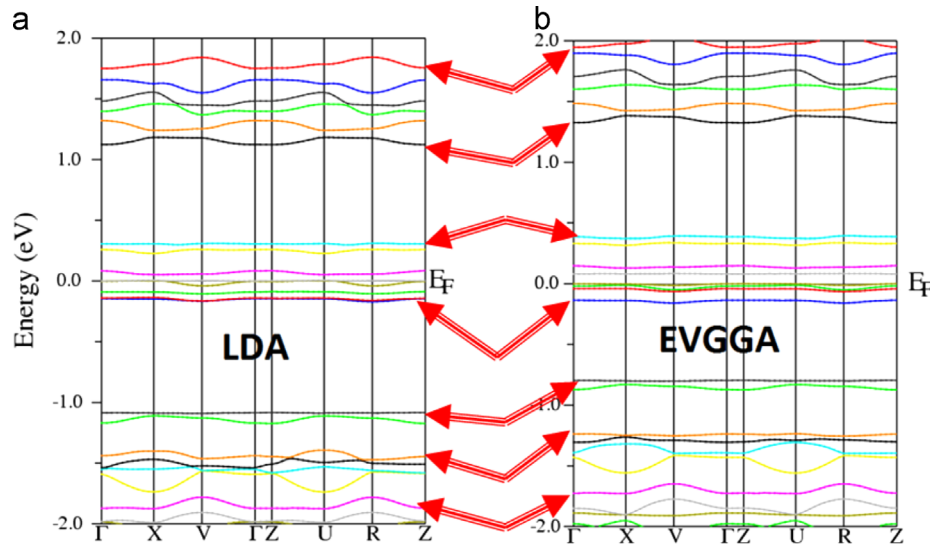


Fig. 3. The calculated electronic band structure; (a) LDA and (b) EVGGA. The zero energy is arbitrarily taken at the Fermi level. The arrows pointed to the differences in the band splitting between LD and EVGGA.

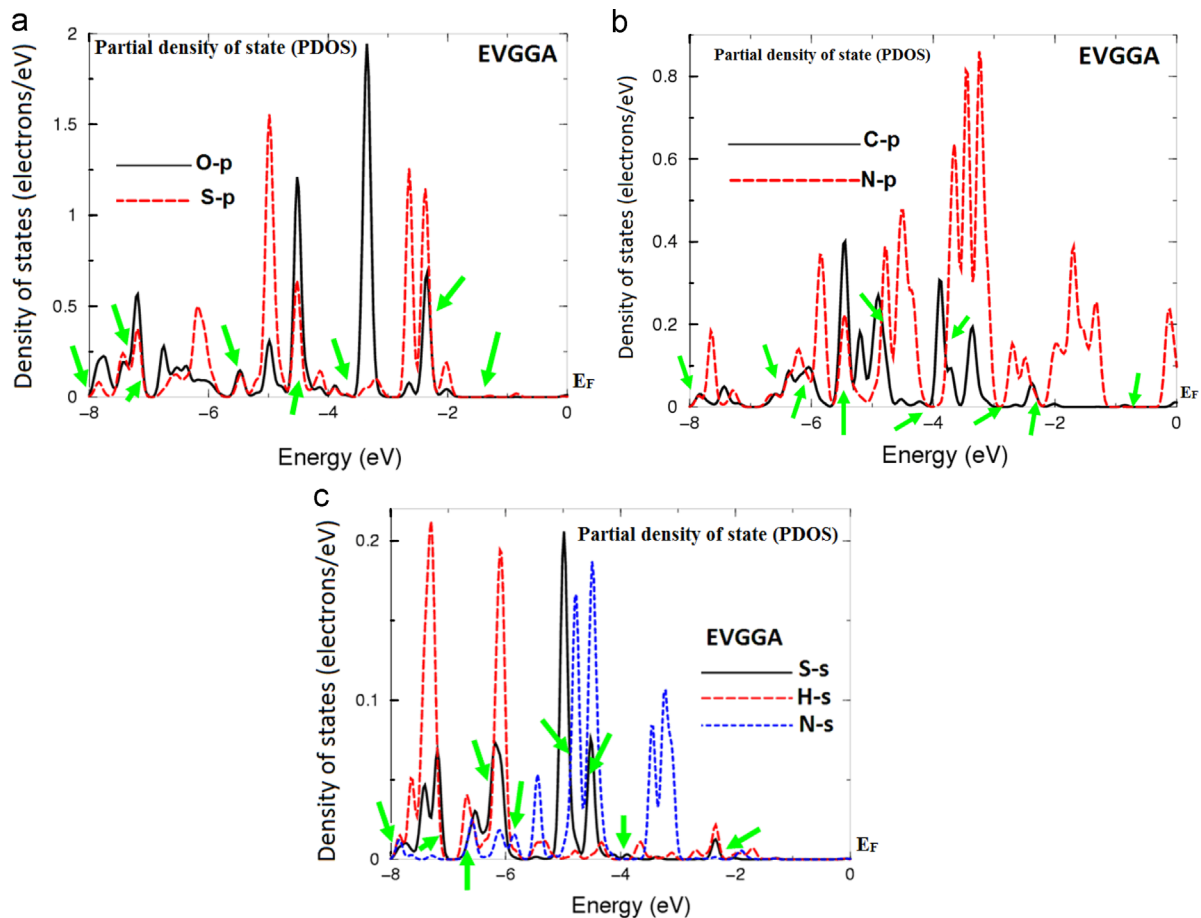


Fig. 4. The angular momentum decomposition of the atoms projected density of states. The zero energy is arbitrarily taken at the Fermi level.

around 0.024 eV resulting in a DOS at $E_F - N(E_F) = 5.50$ states/Ry cell, for LDA and 3.45 states/Ry cell for EVGGA. It is well known that EVGGA produces better band splitting than LDA. Thus, we have enlarged the electronic band structure near E_F in order to show the influences of using LDA and EVGGA on the overlapping of the bands around E_F (see Fig. 3). We, therefore, show all the results for EVGGA approach. These figures suggest that the investigated compound is metallic. We found that N-p states, around E_F , hybridize with S-p, C-s/p and O-p states. The DOS at Fermi energy (E_F) is determined by the overlap between the valence and conduction bands. This overlap indicates a metallic behavior. It is clear that the dispersion of the electronic band structure and DOS obtained within LDA and EVGGA exhibit some differences in the matter of groups/structures which can lead to different electronic properties. The electronic specific heat coefficient (γ),

which is a function of density of states, can be calculated using the expression,

$$\gamma = \frac{1}{3} \pi^2 N(E_F) k_B^2, \quad (1)$$

where $N(E_F)$ is the density of states at Fermi energy E_F , and k_B is the Boltzmann constant. The calculated density of states at Fermi energy $N(E_F)$ enables us to calculate the bare electronic specific heat coefficient, which is found to be 0.95 mJ/mole-K² for LDA and 0.59 mJ/mole-K² for EVGGA. We hope that future experiments will help in determining which of the approximations (GGA or EVGGA) is more suitable.

A remarkable fact regarding the titled compound is that the unit cell contains two independent molecules (A and B) each molecule formed by thirty five atoms. In order to understand the contribution of each orbital in these atoms we investigate the

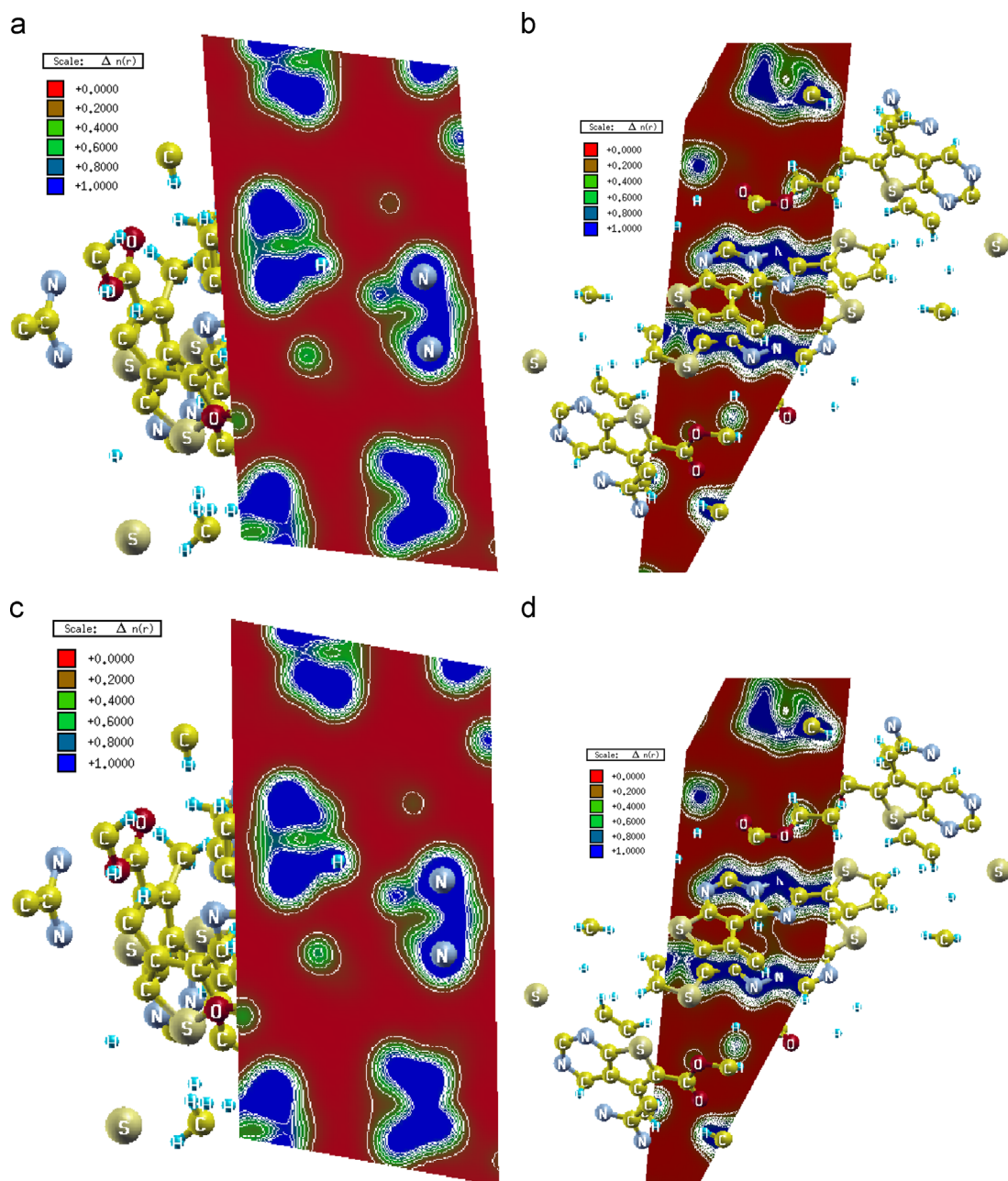


Fig. 5. Electronic space charge density distribution contour in the; (a) (1 0 0) plane LDA; (b) (1 1 0) plane LDA; (c) (1 0 0) EVGGA and (d) (1 1 0) EVGGA.

angular momentum decomposition of the atoms projected density of states (PDOS). Seeking deeper insight picture of the electronic band structure we show the atom-projected PDOS for A and B molecules separately (See [Supplementary material](#) given in Ref. [30]). The atom-projected PDOS helps to identify the angular momentum character of the various structures; the lowest structure from -12.0 eV up to -8.0 eV arises mainly from N-p, O-s/p, C-p states with small contributions from O-p, S-s, C-s, H-s and N-s states. Next structure between -4.0 eV till Fermi level (E_F) originates from O-p, S-p, N-p states with small contribution of O-p, S-s, C-s, H-s and N-s states. Finally the structure from E_F and above is formed mainly from N-p, S-p, C-p with admixture of O-s, S-s, C-s, H-s and N-s states. Also one can see there is a hybridization between various states. At -10.0 eV, N-p, S-p, N-s, S-s, C-p, and O-s states are strongly hybridized. Around the energy of -8.0 eV and -5.0 eV, there exists a strong hybridization between S-p, O-p, N-p, O-s, C-p, H-s and S-s states. N-p, O-p, C-p and N-s states are strongly hybridized around -4.0 eV. At $+2.0$ eV the states S-p hybridized with O-p and N-p. N-s and S-s hybridize around $+4.0$ eV. From $+4.0$ eV and above S-p strongly hybridized with O-p and N-p.

Using the angular momentum decomposition of the atoms projected density of states one can elucidate the origin of the chemical bonding. From [Fig. 4](#), it can be seen that the densities of states, extending from -8.0 eV up to E_F is mainly originated from O-p states (2.0 electrons/eV), S-p states (1.6 electrons/eV), N-p states (0.8 electrons/eV), C-p states (0.4 electrons/eV), H-s states (0.2 electrons/eV), S-s states (0.2 electrons/eV) and N-s states (0.19 electrons/eV). This is obtained by taken very close looking at the angular momentum projected densities of states of O-p, S-p, N-s/p, C-p, H-s, and S-s states as shown in [Fig. 4](#). Following [Fig. 4](#), a, b, and c, it is clear that in the valence band there exists strong and weak hybridization between S-p, O-p, N-s/p, O-s, C-p, H-s and S-s states. The strong hybridizations are pointed by small arrows directly in the figures.

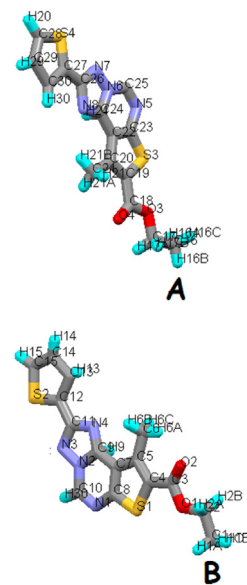
These results show that some electrons from O-p, S-p, N-s/p, C-p, H-s, and S-s states were transferred into valence bands (VBs) and contribute to weak covalence interactions between O–O, S–S and H–H atoms, and substantial covalence interactions between O and C, O and H, N and C, C and S, N and N, C and C and C and H atoms. Therefore, we can also say that the covalent strength of C–H, C–S, C–N, C–O, N–N, C–C and O–H bonds is stronger than that of O–O, S–S and H–H bonds. The interaction due to the strong hybridization and the covalent bond is defined by the degree of hybridization. Hence, there is a strong covalent bonding between the atoms which exhibit strong hybridization.

3.2. Electronic space charge density distribution

The electronic charge density contours in the (1 0 0) and (1 1 0) crystallographic planes of the title compound were investigated to analyze the origin of chemical bonds between all atoms H–O, C–H, N–O, C–O, C–N, C–C and N–N. This is shown in [Fig. 5](#). It is found that majority of charges are accumulated on O and N site and the distribution of electronic charge is non-spherical which results in the bonding between O–H and between H–N showing prevalingly ionic features because the large electro-negativity difference between O (3.44), H (2.20) and N (3.04). Also O forms ionic bonding with C (Pauling electro-negativity=2.55) and covalent bonding with N (3.04). H forms ionic bonding with S and C due to small electro-negativity difference and also due to the hybridized C-p states with H-s states and S-s with H-s states. Moreover, this plot shows that valence electrons from H and S sites are transferred to O, C and N site. This can be seen easily by color charge density scale where blue color (+1.00) corresponds to the maximum charge accumulation site. We find that the choice of the

Table 1
Selected bond lengths (Å) for 9-Methyl-3-Thiophen-2-Yl-Thieno [3,2-e] [1, 2, 4] Thiazolo [4,3-c] Pyrimidine-8-Carboxylic Acid Ethyl Ester, in comparison with experimental data [1].

Bond lengths	Exp.	Theory
C1–C2	1.494(7)/1.480(9)	1.469/1.451
C2–O1	1.461(6)/1.457(6)	1.431/1.425
C3–O2	1.202(6)/1.196(6)	1.180/1.101
C3–O1	1.330(6)/1.325(6)	1.291/1.210
C3–C4	1.475(7)/1.479(6)	1.366/1.372
C4–C5	1.378(6)/1.370(7)	1.265/1.259
C4–S1	1.737(4)/1.736(5)	1.605/1.603
C5–C7	1.417(6)/1.437(6)	1.306/1.318
C5–C6	1.502(6)/1.494(7)	1.410/1.402
C7–C8	1.395(6)/1.389(6)	1.310/1.299
C7–C9	1.427(6)/1.422(6)	1.389/1.387
C8–N1	1.387(6)/1.384(6)	1.297/1.301
C8–S1	1.704(5)/1.722(5)	1.686/1.699
C9–N4	1.328(6)/1.325(6)	1.287/1.267
C9–N2	1.381(6)/1.383(5)	1.301/1.305
C10–N1	1.280(6)/1.283(7)	1.210/1.213
C10–N2	1.374(6)/1.361(7)	1.331/1.322
C11–N3	1.345(6)/1.328(6)	1.299/1.280
C11–N4	1.364(6)/1.368(6)	1.301/1.305
C11–C12	1.452(6)/1.447(6)	1.412/1.401
C12–C15	1.443(8)/1.588(8)	1.408/1.509
C12–S2	1.713(5)/1.706(5)	1.698/1.677
C13–C14	1.454(9)/1.343(7)	1.401/1.299
C14–C15	1.338(8)/1.430(9)	1.299/1.389
C15–S2	1.679(6)/1.663(6)	1.601/1.599
N2–N3	1.374(5)/1.375(5)	1.302/1.300



exchange correlation potential does not effect the bond lengths, angles and the space charge density distribution and hence the type of bonding. A remarkable finding is that there are two independent molecules (A and B) in the asymmetric unit which exhibit intramolecular C–H...O, C–H...N interactions. This intramolecular interaction is different in molecules A and B, where A molecule shows C–H...O interaction while B molecule exhibit C–H...N interaction. We should emphasis that there is π – π interaction between the pyrimidine rings of the two neighbors B molecules which gives extra strengths and stabilizations to the supermolecular structure [1]. The calculated distance between the two neighbors pyrimidine rings was found to be 3.345 Å, in good agreement with the measured one (3.424(1) Å) [1]. From the relaxed geometry we have calculated the bond lengths and the bond angles. The calculated bond lengths and angles were compared with the experimental data [1] and good agreement was found (see [Tables 1 and 2](#)).

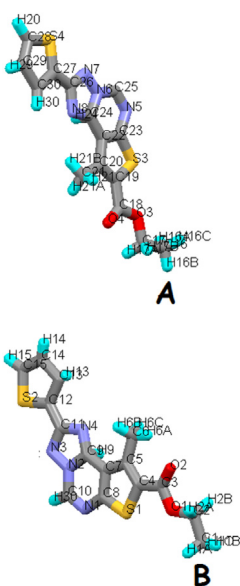
4. Conclusions

The electronic crystal structure, chemical bonding properties, and the electron charge densities of centrosymmetrically 9-Methyl-3-Thiophen-2-Yl-Thieno [3,2-e] [1, 2, 4] Thiazolo [4,3-c] Pyrimidine-8-Carboxylic Acid Ethyl Ester crystals were investigated by the means of density functional theory. The electronic band structure and density of states reveal that; (i) the Engel–Vosko generalized gradient approximation causes to shift Fermi energy towards lower energy by around 0.024 eV. (ii) The density of state at Fermi energy is 5.50 (3.45) states/Ry cell, for local density approximation (Engel–Vosko generalized gradient approximation). (iii) The N-p states, around E_F , overlap with a small admixture of S-p, C-s/p and O-p states. (iv) The calculated bare electronic specific heat coefficient, found to be 0.95 mJ/mole-K² for local density approximation and 0.59 mJ/mole-K² for Engel–

Table 2

Selected bond angles (deg) for 9-Methyl-3-Thiophen-2-Yl-Thieno [3,2-e], [1, 2, 4] Thiazolo [4,3-c] Pyrimidine-8-Carboxylic Acid Ethyl Ester, in comparison with experimental data [1].

Bond angles	Exp.	Theory
O1–C2–C1	107.3(4)/108.2(5)	106.8/1.079
O2–C3–O1	124.1(5)/124.0(4)	123.7/123.5
O2–C3–C4	124.9(4)/124.2(5)	124.0/123.9
O1–C3–C4	111.0(4)/111.7(4)	110.7/110.9
C5–C4–C3	127.8(4)/127.3(4)	127.0/126.8
C5–C4–S1	113.8(3)/114.3(3)	113.1/113.8
C3–C4–S1	118.3(3)/118.4(4)	117.8/117.9
C4–C5–C7	110.1(4)/110.0(4)	109.7/109.5
C4–C5–C6	126.9(4)/126.8(4)	126.1/125.9
C7–C5–C6	123.1(4)/123.1(4)	122.8/122.8
C8–C7–C5	113.4(4)/113.2(4)	112.7/112.5
C8–C7–C9	115.4(4)/115.6(4)	114.8/115.0
N1–C8–C7	126.0(4)/127.0(4)	125.5/126.6
N1–C8–S1	121.7(3)/120.8(3)	121.0/120.1
C7–C8–S1	112.3(3)/112.2(3)	111.8/111.6
N4–C9–N2	109.6(4)/109.9(4)	109.0/109.2
N4–C9–C7	134.3(4)/135.0(4)	129.9/130.8
N2–C9–C7	116.0(4)/115.1(4)	115.8/114.7
N1–C10–N2	122.2(4)/123.1(5)	121.5/122.7
N3–C11–N4	115.9(4)/116.2(4)	115.1/115.8
N3–C11–C12	122.8(4)/122.0(4)	122.0/121.6
N4–C11–C12	121.3(4)/121.8(4)	120.0/121.0
C11–C12–S2	122.8(3)/122.0(4)	122.0/121.4
C13–C12–S2	113.1(4)/118.8(4)	112.7/118.1
C12–C13–C14	106.9(6)/107.9(6)	106.0/107.1
C15–C14–C13	114.2(6)/116.3(6)	113.7/115.7
C14–C15–S2	114.4(5)/114.7(5)	113.9/114.0
C3–O1–C2	116.0(4)/116.7(4)	115.4/116.0
C10–N1–C8	116.5(4)/114.8(4)	115.9/114.0
C10–N2–N3	126.1(4)/126.4(4)	125.5/125.8
C10–N2–C9	123.9(4)/124.4(4)	123.1/123.8
C11–N3–N2	101.3(4)/101.9(3)	100.8/101.0
C9–N4–C11	103.1(4)/102.8(4)	102.7/102.1
C8–S1–C4	90.5(2)/90.3(2)	90.0/89.7
C15–S2–C12	91.4(3)/91.9(3)	89.7/91.1



Vosko generalized gradient approximation. The title compound consists of two independent molecules (A and B) which exhibit different intramolecular interactions, C–H...O interaction in molecule A and C–H...N interaction in molecule B. The π – π interaction between the pyrimidine rings of the two neighbor B molecules gives extra strengths and stabilizations to the supermolecular structure. We find that using different xc potential do not effect the bond lengths, angles and the space charge density distribution and hence the type of the chemical bonding. The calculated lengths and angles of the chemical bonding give deeper insight picture of the electronic structure.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at <http://dx.doi.org/10.1016/j.jmmm.2014.02.067>.

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