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Study on Electronic Properties of α-Uranium using Ab-initio Approach

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ABSTRACT

In the present work, a density functional theory (DFT) based simulation is performed to study the electronic properties of (-uranium through the electronic band structure, total electron density of states (DOS), integrated density of states (IDOS), partial density of states (PDOS) and Fermi -surface measurements. A Generalized Gradient Approximation (GGA) with Projector-Augmented Wave (PAW) pseudopotential is used in present computation. The hybridization between the 5f orbital and 6d orbital also has fairly influences on the electronic Properties of α-Uranium.

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1. Introduction

Uranium is a typical member of the early actinide metallic elements due to its very wide range of applications in the aerospace, military weapons and nuclear industries [1-13]. In general, the α-uranium occurs under normal pressure conditions [3]. Many of the results related to their physical properties have been reported so far by different researchers [1-13]. In general, 5-electron-electron correlations in particular play a crucial role Actinide series [10] for several years. Large volume expansion upon phase transformation is observed [10]. Therefore, theoretical modulations act an important role for studying the surface properties of the materials. Vohra and Spencer [14] have recently observed that at pressures above 116 GPa titanium converts from the hexagonal omega (C32) phase to α-phase.

While, such structure was also studied by Wentzcovitch and Cohen [15] as a probable pathway for the pressure-induced conversion of Magnesium from the hcp (A3) to the bcc (A2) phase. The α-uranium crystal is having an orthorhombic structure at lower 940K temperature and above it, such phase converts to the β-phase and then it converts to the γ-phase at the temperature above 1050K. The α-uranium is stated to be stable up to 70 GPa pressure and at lesser temperatures, while at higher temperatures, the γ-uranium can stabilize its bcc structure [16]. Now a days, uranium is used to power commercial nuclear reactors that produce electricity and to produce isotopes used for medical, industrial, and defence purposes around the world. The study of electronic properties shows the atomic arrangement in such material and from that one can develop or study its isotopic substances for their various nuclear applications.

2. Theory

In condensed matter physics, the characterization of various materials depends on the computational and theoretical techniques. Generally, the Density Functional Theory (DFT) is one of the foremost techniques based on the first principle. The time independent Schrödinger equation for many body systems like solids, nanomaterials and other complex systems can be solved efficiently with the help of DFT [17-26]. For solving the many body interactive systems, wave function method, viz. Hartree and Hartree-Fock, based approximations are utilized. In 1964, Hohenberg and Kohn [27] gave the basis of the theory by saying that, all the ground state properties of a system are the special functional of the density of the ground state interacting system. This reduces the degrees of freedom of a system of N interacting particles from 3N to three coordinates. The limits faced by the theory are that the density is either constant or it varies slowly; however, the actual electronic systems do not fall in any of these categories. In 1965, Kohn and Sham [28] came up with a way out for these shortcomings and gave solution of many body Schrödinger equation in the form of density. The exchange correlation effects fulfilling the Pauli‟s principle, also portraying the coulomb potential are added to the solution, giving results using DFT. Local Density Approximation (LDA) and Generalized Gradient Approximation (GGA) are such approximation that improves the results compared to the conventional wave function method such as Hartree [29] and Hartree-Fock approximations [30]. Various software packages have been developed on the basis of DFT, for the computation of properties of material requiring only information of electron density and some basic crystallographic information of the material. Thus, it is parameter free and hence called first principle theory.

There are several computational codes are available based on the DFT method. A few of them are Quantum ESPRESSO (Quantum open Source Package for Research in Electronic Structure, Simulation and Optimization) [31], SIESTA (Spanish Initiative for Electronic Simulations with Thousands of Atoms) [32], VASP (Vienna Ab-initio Simulation Package) [33], Wien2K [34], CASTEP (Cambridge Serial Total Energy Package) [35] and Abinit [36] etc. These codes have different implementation techniques for the calculation of properties and can be used to complement each other.The density functional theory (DFT) based formulation are found more useful for computing the structural and electronic properties of materials [17-26]. With the advantage of the electronic structure simulation based on density function theory (DFT), the present work mainly focuses on the electronic properties viz; electronic band structure, total electron density of states (DOS), partial DOS, integrated DOS and the Fermi surface study of α-Uranium metal. Such simulations are carried

out through Quantum Espresso package [31] working in WINMOSTAR [37] environment. The GGA with PBE function are utilized to indulge the exchange correlation effects [38] along with the PAW-type pseudopotential [39].

3. Results and Discussion

The plane waves are taken with kinetic energy cut off 20 Ry and Charge density cut off 80 Ry are considered. While, the k-point mesh of $4 \times 4 \times 4$ are used to carried out the integration over the first Brillouin Zone. The equilibrium geometry is seen by optimizing the atomic positions of the said structure and is converged at accuracy in energy of the order of 10-6 Ry. The space group of α-Uranium is Cmcm. In which, total 4-atoms are in its conventional unit cell, with an advent of fcc crystals as displayed in Figure 1.

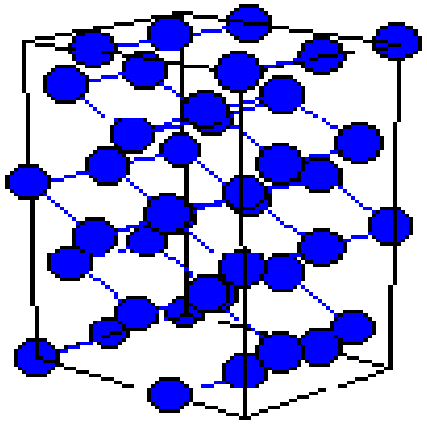

Fig. 1: Crystal Structure of α-Uranium.

Its results from extending a cubic lattice along with two of its orthogonal pairs with a dissimilar lattice parameter. The experimentally lattice parameters are given as $a = 2.84$Å, $b = 5.86$Å and $c = 4.93$ Å [6]. Also, all three vectors intersect at 90° angles (i.e. $\alpha = \beta = \gamma = 90$, hence such three lattice vectors remain mutually orthogonal to each other. The first Brillouin zone of α-Uranium is seen in Figure 2.

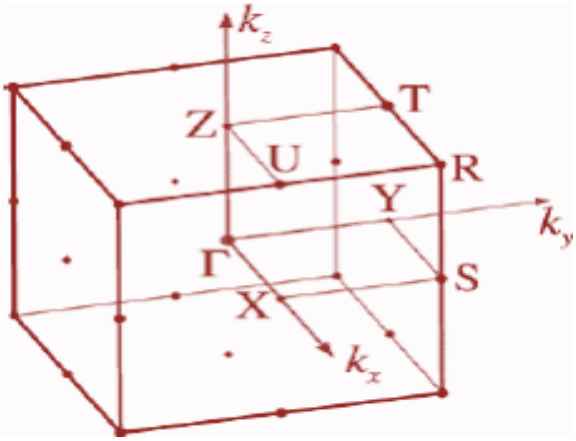

Fig. 2: The first Brillouin zone of α-Uranium.

The electronic band structure, the total electron density of states (DOS), integrated DOS and the partial DOS are displayed Figures. 3-6, respectively for α-Uranium. Here, from Figure 3, it can be observed that the several lowest bands lower the Fermi level nearer to -1 eV are mostly due to f-electrons of the element. The area around Fermi level is formed hence near the Fermi level all bands are dominated. While, in the region above the Fermi level, the contribution of s- and p-electrons is dominant. The band structure was generated for major symmetry points along the path $\Gamma \rightarrow X \rightarrow S \rightarrow R \rightarrow A \rightarrow Z \rightarrow \Gamma \rightarrow Y$. From the nature of the DOS, show deep considerate to examine atomic bonding amongst α-Uranium molecules by the electronic distributions. Figure 4 shows that the s- and p-electrons profound into the atomic core of α-Uranium, which is not observed as valence electrons. Hence, such distribution shows that the typical PAW potential in Quantum Espresso with up to $5s$ and $5p$ electrons is adequate. Besides, the $5f$-electrons governs near the Fermi energy E_{Fermi} amongst all the electrons.

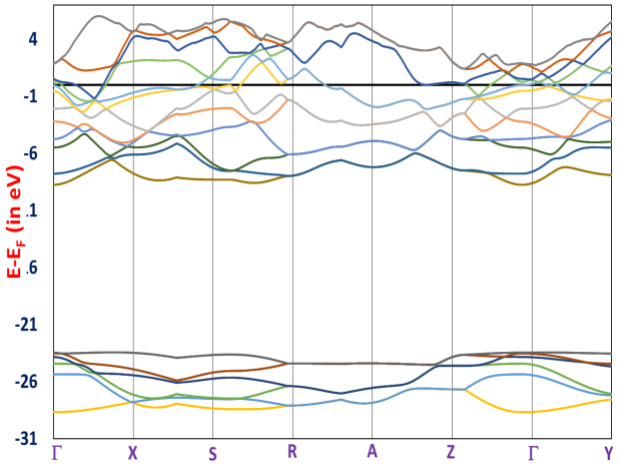

Fig. 3: Electronic band structure of α-Uranium.

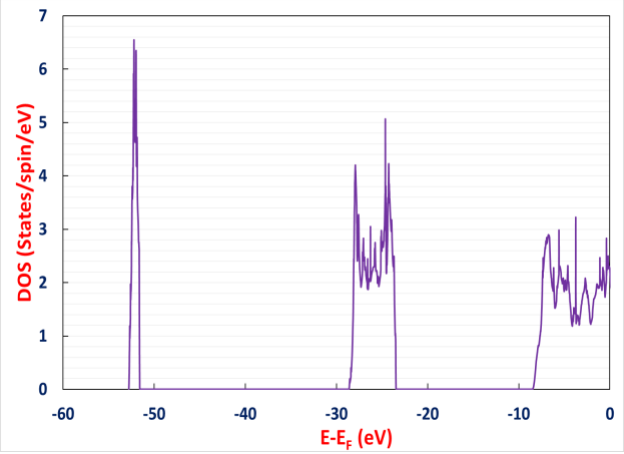

Fig. 4: Total Density of States of α-Uranium.

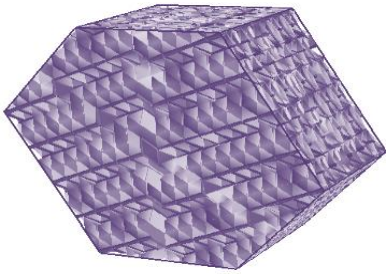

Fig. 7: Fermi surface of α-Uranium.

5. Conclusion

In conclusion, we studied the electronic band structure, DOS, partial DOS, integrated DOS and the Fermi surface of the α-Uranium using the GGA approach with PAW pseudopotential under DFT environment. The dominance of $5f$-state is observed nearby the Fermi level. From the overlapping of the energy bands in the band structure near the E_F, the metallic character of α-Uranium is confirmed.

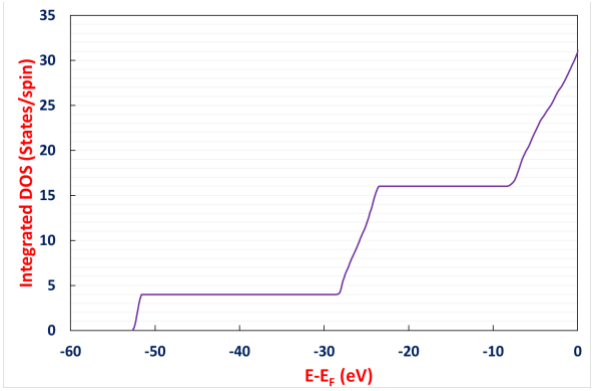

Fig. 5: Integrated Density of States of α-Uranium.

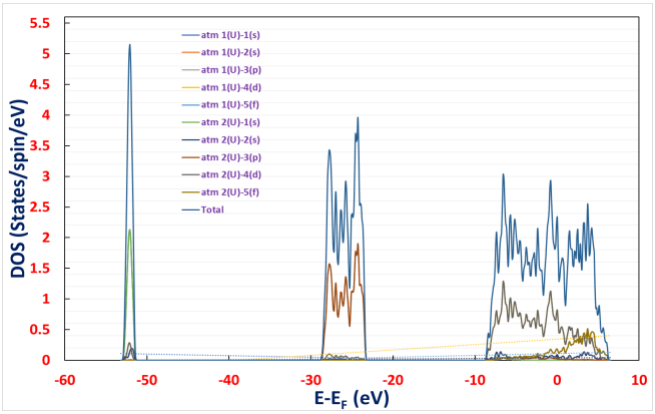

Fig. 6: Partial Density of States of α-Uranium.

The computed Fermi surface is seen in Figure 7. It is because of the restructuring from the charge density waves, the lattice deviates at particular sites and higher sensitivity of the computational methodology.

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