Effect of Roughness on Surface Magnetism of Fe (001)

Priyadarshini Parida, Biplab Ganguli

Abstract— Electronic and magnetic properties of seven layers of bcc Fe are carried out using Augmented Space Recursion (ASR) technique together with tight binding linear muffin tin orbital (TB-LMTO) method. A relativistic self-consistent calculation within local spin density approximation (LSDA) is adopted to generate potential parameters from TB-LMTO Hamiltonian. These parameters are taken as input for ASR calculations. The surface is poisoned with different amount of roughness with random substitution of empty spheres, while the layers beneath the surface remain smooth. Effects of roughness on surface electronic and magnetic properties are studied here. Spin resolved density of states show the effect of roughness on the surface. For all concentration of roughness, magnetic moment of the surface layer is higher than its bulk value. Surface magnetic moment increases with increase in surface roughness.

Index Terms — DFT, TB-LMTO, ASR, Recursion, Magnetic Property, Electronic Property, Surface.

1 Introduction

When a surface is formed, atoms of the topmost layer have looser bonding, lower coordination number and reduced symmetry than that of bulk. Hence some amount of roughness is always present on the surface atomic layer. This roughness is due to interaction of surface atomic layer with the environment above the surface. Such roughness causes significant change in electronic and magnetic properties of surface atoms. Since physical properties of a system depends on detailed electronic structure, atomic arrangement and composition, therefore, it is important to include effect of roughness in electronic structure to study accurate surface properties. In transition metals like Fe, magnetic ordering arises due to the interaction between itinerant fermi electrons. Therefore, in such metals effects of roughness in surface magnetization come via effect of roughness in electronic properties of itinerant electrons as well.

The present study deals with Fe (001) surface with seven atomic layers. Generally bulk properties show after five layers. Therefore, it is important to consider more than five layers to generate a real system. The effects of various amount of roughness on the surface are also carried out using Augmented Space formalism (ASF) [1],[2] together with recursion method [3] and Tight-binding linear muffin tin orbital (TB-LMTO) method [4] within local spin density approximation (LSDA). We have considered two layers of empty spheres on top of the seven layers of Fe to take care of charge leakage.

Calculations based on LCAO [5], FPLAPW [6-7], FPLMTO+SO [8], TB-LMTO Green's function technique [9], TB-LMTO supercell calculation [10],[11], TB-LMTO-ASR [10], TB-LMTO-recursion [11],[12], have shown that surface magnetic moment of Fe is enhanced compared to its bulk value. Most of the above calculations considered smooth surface. The issue of roughness is taken in the work of TB-LMTO recursion method [11],[12]. In this real space technique, the surface roughness is accounted either for a particular environment [12] or in the local curvature of the surface using a stochastic equation and roughness is measured by the scaling exponent [11]. But we have considered all possible configurations and taken average over all for a particular roughness. We introduce roughness by randomly substituting surface Fe atoms by empty spheres. On the surface, with decrease in coordination number, width of surface density of states (DOS) decreases and magnetic moment increases. As we move towards inner atomic layer of bcc Fe, after five layers, the magnetic moment is same as that of bulk value.

2 Computational Methods

ASF is a real space method [1],[2]. It deals with configuration averaging of the random variables by augmenting the Hilbert space spanned by the orbitals with configuration space spanned by the different realizations of the random Hamiltonian.

Recursion method is most suitable to study the electronic properties when translational symmetry is broken, such as surfaces and interfaces. In this method, successive recursion of any initial vector $|u_0\rangle$ on the Hamiltonian gives new vectors, $|u_i\rangle$, which are mutually orthogonal to each other. The recurrence relation is given by,

$$Hu_n = a_n u_n + b_{n+1} u_{n+1} + b_n u_{n-1}$$

a_n and b_n are the recursion coefficient which generate the new tridiagonal Hamiltonian (H). Using this method, the resolvent $(zI-H)^{-1}$ can be written in the form of a continued fraction. The continued fraction is terminated after a finite number of steps using a suitable terminator [13].

ASF with recursion method is very suitable to study the localized properties of disordered systems considering all the configurations. But recursion works on a localized basis. Therefore, it can be combined with TB-LMTO formalism. The full TB-LMTO-ASF Hamiltonian [14] is given by,

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$$H^{\alpha}_{RL,R'L'} = \hat{C}_{RL}\delta_{RR'}\delta_{LL'} + \hat{\Delta}_{RL}S^{\alpha}_{RL,R'L'}\hat{\Delta}_{R'L'}$$
$$\hat{C}_{RL} = C^{A}_{RL}n_R + C^{B}_{RL}(1-n_R)$$
$$\hat{\Delta}_{RL} = \Delta^{A}_{RL}n_R + \Delta^{B}_{RL}(1-n_R)$$

Here R is the lattice sites and $L=(lm)$ are the orbitals indices. For transition metal $l<2$. $C_{RL}{}^{A}$, $C_{RL}{}^{B}$, $\Delta_{RL}{}^{A}$ and $\Delta_{RL}{}^{B}$ are the potential parameters of the constituents A and B of the alloy. n_R are the local site occupation variables which randomly takes value 1 and 0, according to whether the site is occupied by an A atom or not. This method is applied to set up Hamiltonian for interface of two different surfaces such that atoms from each surface diffuse into each other [10]. Therefore, such interface is treated as a disordered system. In our present study, we applied this method to treat material surface and vacuum above as interface. Therefore, one of the types of atom is taken as empty sphere. We have neglected the short-ranged ordering in our calculations.

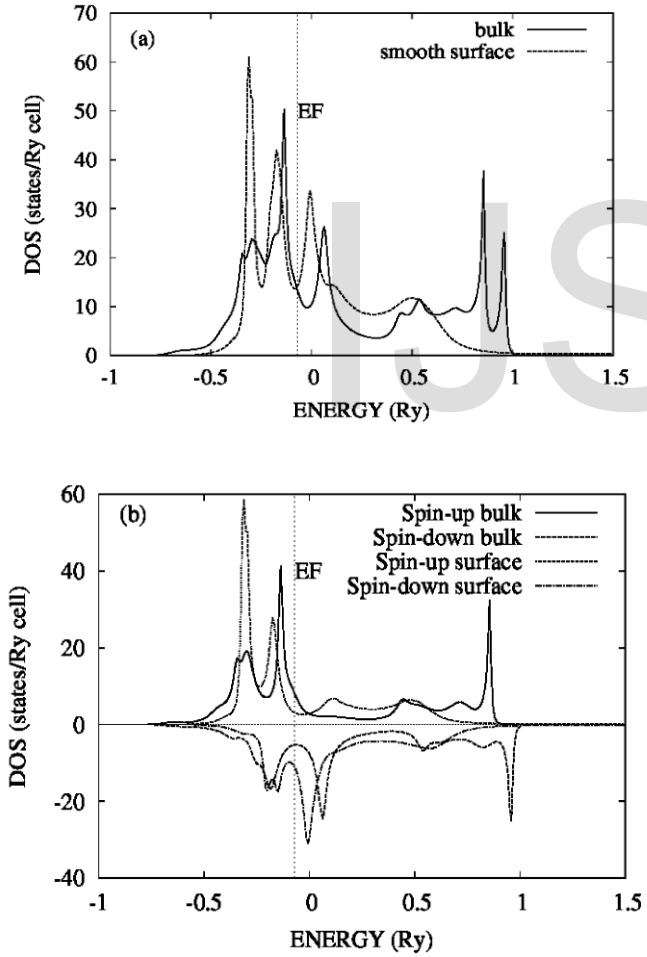


Fig. 1: (a) Total density of states and (b) Spin density of states for smooth surface and bulk bcc Fe (001).

We have chosen seven layers of bcc Fe (001). On top of it, we have introduced two layers of empty spheres which contain charge but no atoms. These empty spheres will take care of the charge leakage into the vacuum. Topmost Fe layer is roughen with various amount of empty spheres ranging from 20% to 80%. The potential parameters are generated from TB-LMTO within LSDA using Barth and Hedin exchange correlation potential. Wave equations are solved by the scalar-relativistic calculations. Potential parameters are different for the topmost layer than the rest. Since topmost layer contains empty spheres, therefore, TB-LMTO is applied to generate parameters for Fe bulk with empty spheres, whereas, for other layers pure Fe bulk is used. Moments of DOS for various layers show convergence with the number of augmented shell. We have used seven shell augmented space calculation and nine steps of recursion. We have used Luchini and Nex terminator [13] to terminate the continued fraction of the recursion method.

3 Results & Discussions

Fig. 1 shows, the width of surface DOS decreases compared to its bulk value for smooth surface. This is due to decrease in nearest neighbor at the surface and looser bonding of the surface atoms. As we move towards the inner atomic layers, nearest neighbor increases and width of DOS increases and it approaches towards bulk DOS as shown in fig. 2. For the surface layer fermi energy is -0.19 Ry. But after five layer, the fermi energy approaches to -0.07 Ry which is close to bulk (-0.08 Ry). The bulk fermi energy is taken from bulk calculation of bcc Fe within TB-LMTO.

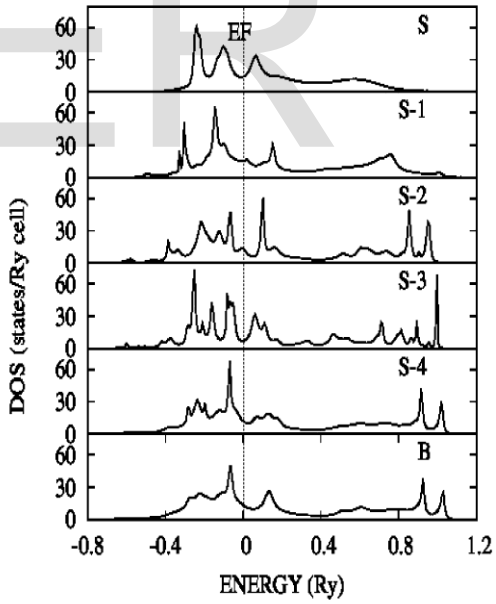
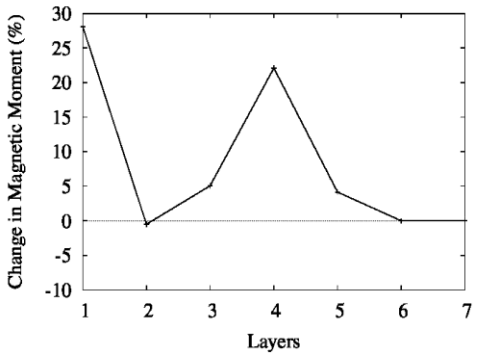


Fig. 2: Total density of states for surface (S), sub-surfaces (S-1, S-2, S-3, S-4) and bulk (B) bcc Fe (001). The vertical line at zero represents Fermi energy.

A comparison on the magnetic moment for the surface layer (S), sub-surfaces layers (S-1 and S-2), and bulk (B) is given in table 1 for smooth surface. We find the surface magnetic moment to be 2.78 μ_B/atom and that of the bulk 2.17 μ_B/atom. Here, the surface magnetic moment is slightly less than the earlier studies. But the bulk magnetic moment is well compa-

rable to FPLAPW study [7], FP-LMTO study [8] and supercell calculations [10]. The surface magnetic moment enhances by 28% than the bulk value. For the sub-surface layer (S-1), our calculated magnetic moment is 2.16 μ_B/atom which agrees with supercell calculation [10],[11], TB-LMTO-ASR study [10], TB-LMTO recursion study [11],[12]. Magnetic moment of sub-surface layer (S-1) is same as that of bulk value. Fig. 3 shows, magnetic moment oscillates as we go down the layers. After five layers from the surface, DOS approaches the bulk DOS. Fermi energy is closed to that of bulk value and the magnetic moment is also the same as that of bulk. Therefore, it is assumed that the bulk property is obtained after five layers.



Figue 3: Change in magnetic moment from surface to the bulk. Layer 1 represents the surface.

TABLE 1
MAGNETIC MOMENT IN μ_B/ATOM FOR SURFACE (S), SUB-SURFACES (S-1 AND S-2) and CENTRAL LAYER OR BULK (C/B) FOR BCC Fe (001). NUMBERS IN THE SQUARE BRACKET REPRESENTS THE REFERENCE NUMBERS.

Methods used	S	S-1	S-2	C/B
TB-LMTO Supercell	2.86 [10], 2.98 [11]	2.16 [10], 2.17 [11]	2.38 [10], 2.40 [11]	2.17 [10], 2.26 [11]
TB-LMTO-ASR dialation [10]	2.99	2.17	2.38	2.27
TB-LMTO Recursion	2.99[11], 2.95 [12]	2.21[11], 2.2 [12]	2.38 [11], 2.387[12]	2.26 [11], 2.28 [12]
FPLAPW	2.98 [6,7]	2.35 [6]	2.39 [6]	2.2 [6], 2.15 [7]
FPLMTO+Spin Orbit [8]	2.87	2.34	2.33	2.18
TB-LMTO Green's function [9]	2.97	2.30	2.37	2.24
LCAO [5]	3.01	1.69	2.13	1.84
Experimental				2.21[15]
Our Work	2.78	2.16	2.28	2.17

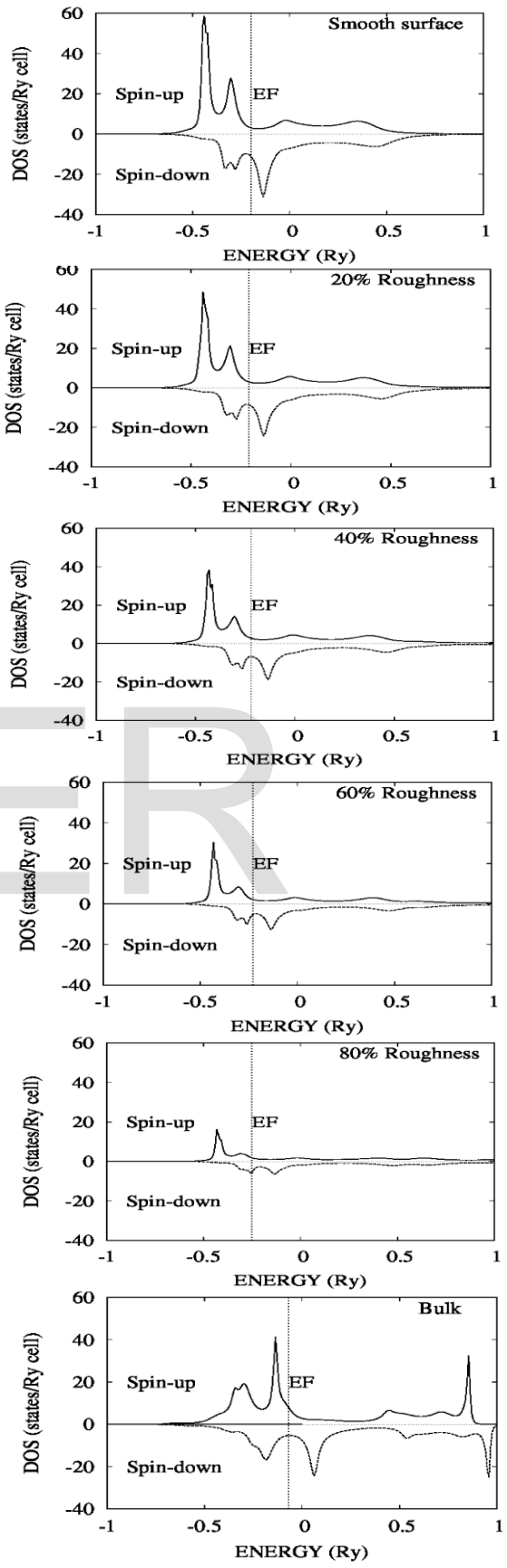


Fig. 4: Spin up and spin down DOS for different roughness.

Fig. 4 shows that with increase in roughness on the topmost layer, fermi energy is shifted towards lower energy value. For smooth surface fermi energy is -0.19 Ry. But for a 80% rough surface, it is -0.25 Ry. With increase in the roughness, width of surface DOS also increases. It is clear from table 2 that magnetic moment increases with roughness. For 80% roughness on the top most Fe layer, the magnetic moment is 3.10 μ_B/atom, whereas, for 20% roughness, the magnetic moment reduces to 2.85 μ_B/atom.

The average magnetic moment (M_{avg}) is calculated using $M_{avg} = (1-x)M_{Fe} + xM_{Es}$. Here, x is the concentration of empty spheres. M_{Fe} and M_{Es} are magnetic moment of Fe and empty spheres respectively. M_{Es} is negligible. Fig. 5 shows, with decrease in roughness, average magnetic moment of surface layer increases. With increase in roughness, there is reduction in the coordination number of Fe atom and hence the interaction among Fe atoms decreases. Due to this, local magnetic moment of Fe atom in surface layer increases.

TABLE 2
VARIATION OF LOCAL MAGNETIC MOMENT OF Fe IN SURFACE LAYER AND AVERAGE MAGNETIC MOMENT OF SURFACE LAYER WITH ROUGHENSS IN μ_B/ATOM

Percentage of Roughness	Magnetic moment of Fe	Average Magnetic moment of surface
80	3.10	0.63
60	2.91	1.17
40	2.86	1.72
20	2.85	2.28
0	2.78	2.78

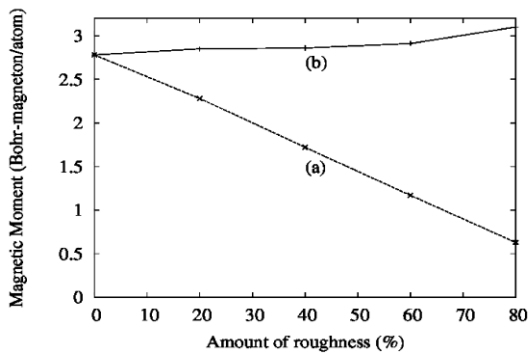


Fig. 5: Variation of (a) average magnetic moment of surface layer and (b) magnetic moment of Fe in surface layer with various amount of roughness.

4 Conclusion

We have carried out TB-LMTO-ASR study to calculate the change in electronic and magnetic properties for seven layer bcc Fe (001) from surface to bulk. The width of DOS decreases at the surface than the bulk for smooth surface. The surface magnetic moment increases by 28% of the bulk value. The topmost layer is roughen with various amount of empty spheres and its effects on surface electronic and magnetic properties are carried out. With increase in roughness the width of the surface DOS increases and also magnetic moment increases.

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