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Electrical and Magnetic Properties of Rough Alloyed Interface of Fe/Co.

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Abstract

We investigated the electronic structure of rough interfaces in random Co/Fe alloys, where cobalt (Co) was deposited on an iron (Fe) substrate with varying composition ratios. To model the rough surface, we utilized a coupled continuum equation as described by Huda. The electronic properties, specifically the density of states of these random alloys, were calculated using the augmented space recursion method in conjunction with the Linear Muffin-tin Orbital (LMTO) Method. Furthermore, we analyzed the magnetic and electronic properties derived from the calculated density of states.

Keywords: Electronic structure, Random alloys, LMTO, Augmented space recursion, Density of States (DOS), Magnetic moments of alloy.

1. Introduction

The study of magnetic thin films and their interfaces is of paramount importance in the field of spintronics, where the control of spin currents in magnetic materials is critical for the development of advanced electronic devices. Among the various magnetic materials, iron (Fe) and cobalt (Co) alloys have garnered significant attention due to their high magnetic moment, robust magnetic anisotropy, and compatibility with existing technology.

Several studies (Fu et al., 1985; Bak et al., 1987; Das Sharma & Kotliar, 1994; Barabasi & Stanley, 1995) have attempted to elucidate the dynamics of surface growth during deposition. A model developed by Sanyal has furthered our understanding of kinetic roughening by using coupled continuum equations for rough surfaces and mobile atoms (Sanyal et al., 1999).

Most of the research on surface growth has focused on the quantification and characterization of surface roughness. Sanyal demonstrated that this characterization can be achieved experimentally (Sanyal, 1998; Sinha et al., 1996), for instance, through grazing-incidence small-angle X-ray scattering experiments. The magnetic moment of an itinerant atom in a solid is inherently a local property, varying randomly across a rough surface (Huda & Mookerjee, 2003). The collective behavior of interacting itinerant electrons influences both the electronic properties and the density of states.

It is also important to note the significant differences in coordination numbers between surface atoms and bulk atoms due to their differing environments. Surface atoms exhibit weaker bonding because they have fewer neighboring atoms compared to bulk atoms. This difference further contributes to the unique electronic and magnetic properties observed at rough surfaces.

2. Methodology

The LMTO method builds upon the properties of muffin-tin orbitals (Skriver, 1984; Andersen, 1975). The LMTO approach has led to many new concepts, theories, and features. The LMTO methodology has developed in a way most appropriate for close-packed solids, and the descriptions in the literature are often difficult to penetrate because the basic theory is interwoven with approximations and motivational aspects. The LMTO method is the linearized version (Andersen, 1975) of the Korringa-Kohn-Rostoker (KKR) method. Computationally, it is one of the fastest of the linear methods and its formalism is extremely simple. It is transparent too. This method is generally used to perform density-functional calculations for crystals and alloys (Skriver, 1984).

The density functional theory (DFT) minimizes the many-body Hamiltonian of the valence electron cloud to an effective one-electron problem. The effective ‘Hamiltonian’ within the LDA is;

$$V(r) = \sum_{R} v(r - R), \tag{2.1}$$

where $'R'$ denotes the position of ion-core and r is the position vector of the electron.

The Schrödinger-like Kohn-Sham equation for the single electron, within the DFT, is given by;

$$\left[-\frac{\hbar^2}{2m}\nabla^2 + v(r - R)\right]\varphi(r - R, \varepsilon) = \varepsilon\, \varphi(r - R, \varepsilon), \tag{2.2}$$

The local environment approach (Heine, 1980) to the electronic structure of solids needs an alternative to the band theory to solve the Schrodinger equation. The Recursion method is an efficient computational method for condensed matter applications. This method was introduced (Haydock *et al*., 1972) as an elegant alternative to the well-established reciprocal space-based methods for calculating electronic and other properties of solids. This is done by constructing a new orthonormal basis set $|n\}$ from the older one $|n\rangle$ by the following three terms recurrence relation:

$$|n + 1\} = H|n\} + \alpha_n|n\} + \beta_n^2|n - 1\}, \tag{2.3}$$

where α_n and β_n are two sets of real parameters obtained by imposing the orthonormalizability condition of the basis. They are given by:

$$\alpha_n = \frac{\{n|H|n\}}{\{n|n\}}, \text{ and} \beta_n^2 = \frac{\{n + 1|H|n\}}{\{n + 1|n + 1\}}.$$

The initial condition of the recursion relation is;

$$|1\} = |RL_\alpha\}, \text{ and} \beta_0^2 = 1.$$

The TB-LMTO (Andersen *et al*., 1984) and Recursion Methods (Haydock *et al*., 1972) are dealing with the study of disordered systems. Besides these two methods, the augmented space method (ASM) (Mookerjee, 1973) is probably one of the appropriate techniques for handling configuration averaging. The Tight-Binding method and Linearized Muffin-tin Orbital Method (TB-LMTO) (Andersen *et al*., 1984) permits us to compute an efficient Hamiltonian from which the electronic structure are often derived with appreciable accuracy and speed. The Hamiltonian is a sparse matrix in real space. This practice is well suited to elucidate the structurally disordered systems. The recursion method also allows us to compute directly in real space and is the right technique to use along with the TB-LMTO method when we are handling with disordered systems.

The regular procedure now is to propose the mean-field approximation: the coherent potential approximation (CPA), the traveling cluster approximation (TCA), various cluster CPAs, and so on. The generalization of the CPA is generally very difficult for practical computational methods. However, we can propose an alternate approach that is both accurate, fast, and which has controllable approximations. We also like to apply the recursion method, a very efficient method described by Haydock (Haydock, *et al*., 1972) directly on the augmented space without carrying out mean-field like approximations.

For a random binary alloys, the first order LMTO Hamiltonian within the most tight-binding representation is:

$$H = \sum_{i} C_i P_i + \sum_{ij} \Delta_i^{-1/2} S_{ij} \Delta_j^{-1/2} T_{ij}, \tag{2.4}$$

With,

$$C_i = C_{i,L}\delta_{L\acute{L}} = C_A x_i + C_B(1 - x_i),$$

$$\Delta_i^{-1/2} = \Delta_{i,L}^{-1/2}\delta_{L\acute{L}} = \Delta_A^{-1/2} x_i + \Delta_B^{-1/2}(1 - x_i),$$

$$S_{ij} = S_{iL,j\acute{L}}. \tag{2.5}$$

where j = i, or the nearest or next-nearest neighbours of *i* on the lattice.

Also, this Hamiltonian can be transformed by little algebra as:

$$H = H_B + \sum_i \delta C\ x_i P_i + \sum_{ij} \left[\Delta^{-1/2} x_i S_{ij} \Delta_B^{-1/2} + \cdots + \Delta_B^{-1/2} S_{ij} \Delta^{-1/2} x_j + \Delta^{-1/2} x_i S_{ij} \Delta^{-1/2} x_j \right], \tag{2.6}$$

With,

$$\delta C = C_B - C_A, \quad \text{and} \Delta^{-1/2} = \Delta_B^{-1/2} - \Delta_A^{-1/2}.$$

Let convert this into the Augmented Space Hamiltonian as follows:

$$\tilde{H} = H_B \otimes \tilde{I} + \sum_i \delta C\ P_i \otimes M^{(i)} + \sum_{ij} \left[\Delta^{-1/2}\ S_{ij} \Delta_B^{-1/2}\ \widehat{T_{ij}} \otimes \widehat{M^{(i)}} + \cdots + \Delta_B^{-1/2} S_{ij} \Delta^{-1/2} \widehat{T_{ij}} \otimes \widehat{M^{(i)}} + \ \Delta^{-1/2} S_{ij} \Delta^{-1/2}\ \widehat{T_{ij}} \otimes \widehat{M^{(i)}} \otimes \widehat{M^{(i)}} \right],$$

Or,

$$\tilde{H} = \tilde{H}_B + \tilde{H}_1 + \tilde{H}_{2a} + \tilde{H}_{2b} + \tilde{H}_3. \tag{2.7}$$

Is is known for us how $\tilde{H}_1$ acts on the members of augmented space basis. There are four distinct types of this Hamiltonian:

- $H^{(0)}$, acts on the real space.
- $H^{(1)}$, acts on the configuration space.
- $H^{(2)}$, acts both on real and configuration space, but it can change the configuration only at one site.
- $H^{(4)}$, acts on both on the real and configuration space and it can change the configuration at two sites.

The configuration store as a binary string. The string store as bits of words. A recursion calculation every time starts with a computation on a finite part of the lattice. That reduces it to the recursive way of storing the configurations.

From the following equation give us the recursion coefficients:

$$|n\} = \tilde{H}|n-1\} - \alpha_{n-1}|n-1\} - \beta_{n-1}^2|n-2\}, \tag{2.8}$$

$$\text{where,}\ \ \alpha_n = \{n\big|\tilde{H}\big|n\},\ \ \beta_n^2 = \{n|n\}/\{n-1|n-1\}, \text{and} |0\} = 0.$$

Suppose we generate these coefficients up-to a finite number of recursion steps, say, p, then we write:

$$\langle G_{ii}(z) \rangle_{av} = \cfrac{1}{z - \alpha_1 - \cfrac{\beta_1^2}{z-\alpha_2 - \cfrac{\beta_2^2}{\ddots \cfrac{\ddots}{z-\alpha_p - T(z)}}}}. \tag{2.9}$$

According to the Haydock procedure first we calculate the coefficients $\{\alpha_n, \beta_n\} n = 1,2,3, \ldots P$, then find the terminator $T(z)$(Haydock, 1995). Mookerjee suggests that in conjunction (Mookerjee, 1995) including the TB-LMTO and Recursion packages, the Augmented Space method certainly replaces difficult and most costly mean-field approaches and its generalizations.

We can define the second order Hamiltonian, which is generated from of self-consistent calculation of TB-LMTO has the form:

$$H^{(\sigma,i)} = E_v^{(\sigma,i)} + h^{(\sigma,i)} - h^{(\sigma,i)} o^{(\sigma,i)} h^{(\sigma,i)}, \tag{2.10}$$

where,

$$h^{(\sigma,i)} = \sum_{RL} \left(C_{RL}^{(\sigma,i)} - E_{v,L}^{(\sigma,i)} \right) P_{RL} + \sum_{RL} \sum_{R'L'} \Delta_{RL}^{1/2(\sigma,i)} S_{RL,R'L'}^{(i)} \Delta_{R'L'}^{1/2(\sigma,i)} T_{RL\alpha,R'L'\alpha'}.$$

The position of the atoms are labelled by R and *i* denotes the layer which are below the surface R, and composite angular momentum represent by $= l, m$, σ the spin index (either ↑ or ↓). $C_{RL}^{(\sigma,i)}$and $O_{RL}^{(\sigma,i)}$ are the potential parameters and

overlapping matrix of the TB-LMTO method. The width parameter is denoted by $\Delta_{RL}^{1/2(\sigma,i)}$ and the short ranged screened structure matrix is $S_{RL,R'L'}^{(i)}$, which depends only on the geometry of underlying lattice. It is observed that the screened structure matrix varies at different position of rough surface because of surface dilation. H denotes the Hilbert space spanned by the tight binding basis {|RL >}. The projection and transfer operators in H are P_{RL} and $T_{RL,R'L'}$.

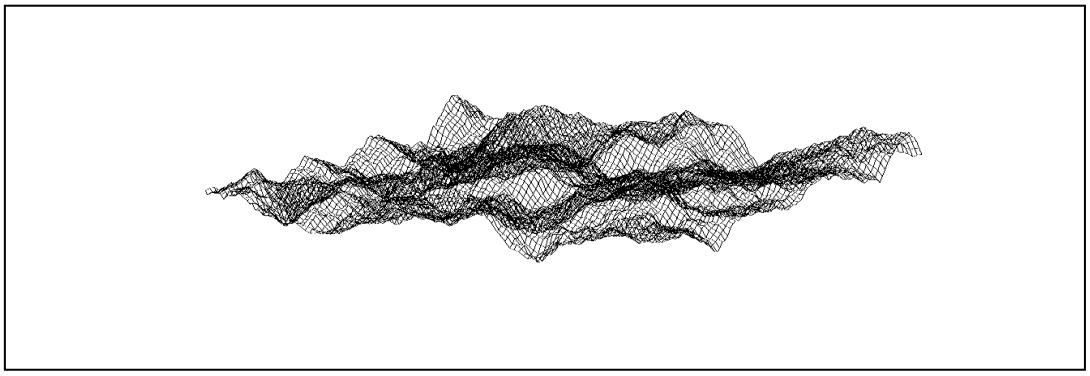

Figure 1. A part of the rough surface generated from coupled continuum equations

The Green's function is defined as,

$$G_{ii}(z) = G_{RL,RL}^{(\sigma,i)} = \langle R, L \left| \left(EI - H^{(\sigma,i}\right)^{-1} \right| R, L \rangle,$$

$$= \frac{1}{E - \alpha_0 - \cfrac{\beta_1^2}{E-\alpha_1 - \cfrac{\beta_2^2}{E-\alpha_2 - \cfrac{\beta_3^2}{\vdots \\ E-\alpha_n - \cfrac{\beta_n^2}{T_n(z)}}}}}, \tag{2.11}$$

The initial part of the continuum fraction gives us an appropriate terminator, $T_n(z)$. This terminator has the Herglotz analytic properties. The density states can be calculated from the imaginary part of the Green function, which again gives us the charge and magnetization densities. The charge and magnetization densities become the input of the self-consistency iterations using the Local Spin Density Approximation (LSDA). The input parameters in (2.10) depend on layer and dissimilar from those of the bulk. We required a few super-cell calculations to start out potential parameters. This was done using the trial and error method by taking a unit cell varying the amount of Fe and empty spheres. Using these parameters, we did a recursion calculation and observed that the density of the (100) plane matches all right with the result [Huda A and Mookerjee M, 2003] obtained from the LMTO.

3. Computational Details

To calculate the electronic and magnetic properties, we require a rough interface, where two different types of atoms are deposited from two sides. In our present study, we used a model that depicts the Molecular Beam Epitaxy method. First of all, we consider the deposition of Fe on a heated substrate and evaporation-accreting-based coupled equation to lead us to a surface shown in Figure: 1. After that, we deposited Co atoms on the surface. We will have a random type alloy in the interface because of the diffusion and segregation of atoms. The percent of atoms in the interface depends on the surface morphology, and it is assumed that the percent of the previously deposited atoms (in present case Fe) is higher in grooves and less in the mounds.

We assume that in the case of a random binary alloys, $A_xB_{1-x,}$ where A is the previously deposited atoms, the value of x should not be lower than 60%. In the present study, we calculated the electronic and magnetic properties of Fe_xCo_{1-x}, where the value of x is 60%, 65%, 70%, 75%, 80%, 85%, 90% and 95%.

We generated a real space cluster consisting of six hundred atoms and an augmented space shell up to the seventh nearest neighbor from the zeroeth (or starting) state was used to find out the projected mean density of states of binary alloy related to the paramagnetic and ferromagnetic materials. We accurately calculated nine pairs of recursion coefficients. The continued fraction which was originated from the Green's function was terminated by an analytic terminator which was defined by Luchini & Nex, (1987). Ghosh et al., (1999) showed the convergence of the Fermi energy, the band energy,

the magnetic moments, and the charge densities, obtain within the augmented space recursion. In our work, the convergence tests were carried out with prescribe accuracies as suggested by the authors. The computational burden of the recursion calculation within the whole augmented space was reduced by utilizing the native symmetries of the augmented space. The seed recursion formula was used with sixteen energy seed points uniformly across the spectrum.

The reduction techniques are represented well within the reference papers and therefore the readers have observed them for details. It is wrong interpreted earlier that though the augmented space recursion is analytically correct but it is not workable for application as a computational technique for real random alloys. Saha et. al., (1996) showed that the augmented space recursion with an analytic terminator always sets off Herglotz results, whether we use the version including short-ranged order (Mookerjee & Prasad, 1993) local lattice distortions (Saha. et. al, 1994) or the homogeneous disorder model as in this paper.

For the atoms Fe and Co we chose the Wigner-Seitz radii in such a fashion that the typical volume occupied by the atoms remains preserved. Using the volume preserved, we varied the radii in order that the ultimate configuration has neutral spheres. In this condition, we do not have to incorporate the averaged Madelung energy part in the whole energy of the alloy. The definition and computation of the Madelung Energy during a random alloy had round-faced contestation and up to the present date, no satisfactory solution of the problem exists. At the same time, we have made that the sphere overlap remain inside the 15% limit advised by Andersen.

The calculations are created self-consistent within the LSDA sense. At every single stage, the averaged charge densities are measured from the augmented space recursion, and also a new potential is computed by the standard LSDA method. The total energy and charges are calculated self-consistently to errors of the order of .00001. The overall energy related to the lattice constant was also minimized. The presented results are for the minimum configuration of the lattice. We have neither taken into account the short-ranged ordering due to chemical clustering nor any lattice distortions due to the size mismatches between the two constituents.

4. Results and Discussion

The ground-state crystalline and magnetic structures of pure Fe and Co are body-centered cubic (bcc) and ferromagnetic, respectively. In the case of bcc FexCo1-x alloys, the ferromagnetic phase remains stable across the entire concentration range. For this study, we focused on regions of the magnetic phase diagram where the ferromagnetic phase is stable, ensuring that our analysis remains relevant to practical applications.

Given that cobalt was deposited on a rough iron substrate, we anticipated that the maximum concentration of Co within the alloy would be approximately 40%. This constraint on cobalt concentration is consistent with the nature of the rough interface, where alloying and surface roughness influence the achievable Co content.

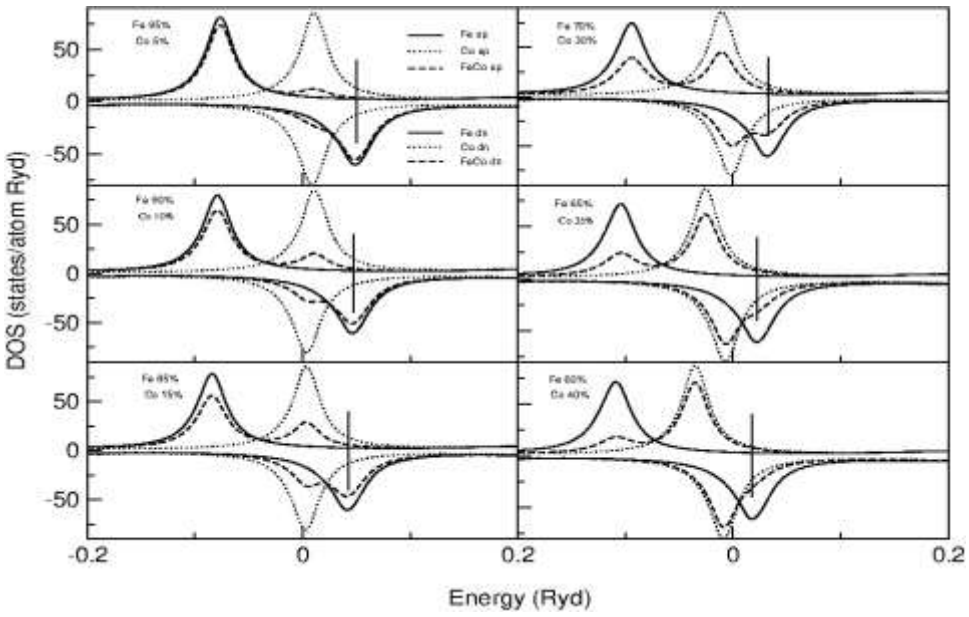

Figure 2. The spin-resolved partial and total density of states of Fe_xCo_{1-x} alloys. We represent only x=95%, 90%, 85%, 80%, 75%, 70%, 65% and 60%

The partial and total density of states of Fe_xCo_{1-x} alloys shows in Figure-2. With the increase of Co in the alloys, we

observe a shift of d-bands to the left. We also observe that Fermi energy is decreasing with increase of Co concentration [Figure: 3].

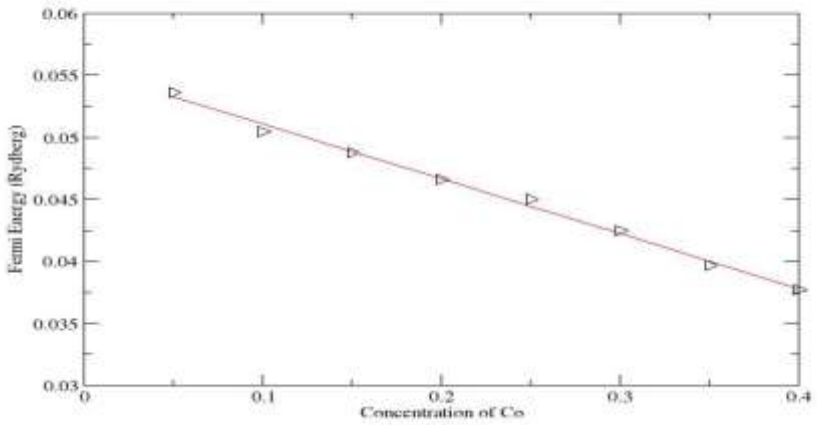

Figure 3. The Fermi energy of Fe_xCo_{1-x} with different values of x

In Figure: 4 represents the magnetic moments of individual atoms and total magnetic moments of the alloys. We found that the magnetic moment of Co is increasing with the increase of Co constituents, but the magnetic moment of Fe is decreasing. The overall magnetic moments of Alloys is decreasing with the increase of Co. The mapping of Curvature and itinerant magnetic moments are still not done. It is assumed that there will be a relation between the local curvature and the magnetic moment.

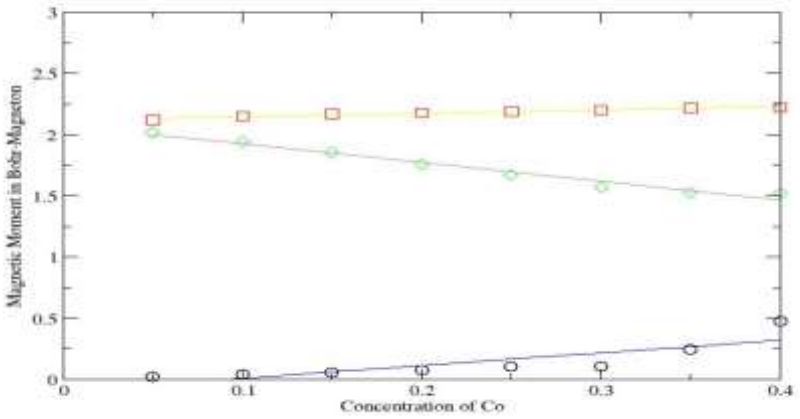

Figure 4. The magnetic moments of Fe, and Co in Fe_xCo_{1-x} defined by squares and circle, respectively. The magnetic moments of $FexCo1_{-x\ alloy}$ are represented by diamond

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