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Surface Relaxation Effect on the Magnetism of Fe Overlayer on Cr (001)

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The effects of surface relaxation on surface and interface magnetism in Fe / Cr (001) are investigated using the highly precise all-electron total-energy full-potential linearized augmented plane wave method. The Fe-Cr interlayer spacing is determined by total-energy calculation and it is found to be relaxed downward by 18 %. For the relaxed system, the magnetic moment of surface Fe is highly suppressed to be 1.72 μ_B compared to the unrelaxed case (2.39 μ_B). This reduction of magnetic moment is considered as a result of the enhanced hybridization between Fe-*d* and Cr-*d* states, which can be seen from the calculated density of states. This work suggests the importance of effect of relaxation to the surface and interface magnetism in Fe / Cr system.

1. Introduction

Magnetism at surfaces and interfaces of transition metals continues to attract considerable experimental and theoretical attention. Recently developed sophisticated synthesis techniques permit the fabrication of a variety of artificial materials such as thin films, sandwiches, and modulated structures. All-electron first principle theoretical approaches have been developed such as full-potential linearized augmented plane wave (FLAPW) method [1] which gives highly precise descriptions of the electronic, magnetic, and structural properties of thin films and bulk solids. The interplay of theory and experiment has yielded interesting results. For example, enhanced magnetism at Fe [2], Ni [3], and Cr [4] surfaces has been predicted and observed.

Victora and Falicov [5] showed that moment of Cr overlayer on Fe is enhanced to be 3.63 μ_B using Slater-Koster parameterized tight-binding (TB) method. Fu *et al.* [6] predicted strongly enhanced two-dimensional ferromagnetic moments on metallic overlayers, interfaces, and superlattices. In their prediction, the interface layer Fe, sandwiched by Cr and Au (001), has a reduced moment of -1.96 μ_B. For Fe overlayer on Cr / Ag (001), the Fe moment is reduced to be 2.3 μ_B, smaller than that of the surface layer of clean Fe (001) [2].

Strong antiferromagnetic (AFM) couplings between Fe layers separated by Cr have been found in Fe / Cr / Fe sandwiches [7] and Fe / Cr superlattices [8]. Xu and Freeman [9] investigated that the electronic structures of bcc Fe_m/Cr_n (001) ($m = 1$, 3 and $n = 1$, 3, 5, 7) superlattices using the self-consistent total-energy linear muffin-tin orbital (LMTO) method with the combined correction term. Their result showed that there is a strong hybridization between Cr-*d* and Fe-*d* states.

Kang *et al.* [10] investigated the electronic structures of Fe on Cr (Fe / Cr) and Cr / Fe / Cr sandwich films, with an Fe coverage of 1-20 Å, by using photoemission spectroscopy (PES). In their results, it is observed a sharp emission just at Fermi energy (E_F) which originated primarily from hybridization between Cr-*d* and Fe-*d* states at the Fe / Cr interface and partially from the Fe-*d* surface states in the Fe overlayer. They also compared their experimental results with supercell band-structure calculations for a system with a monolayer (ML) Fe on each side of 5 ML Cr, Fe (1 ML) / Cr (5 ML) / Fe (1 ML) by LMTO band method. This comparision reveals that the effects of surface states and hybridization between Fe-*d* and Cr-*d* states, both of which contribute to the sharp emission at E_F for a very thin Fe overlayer, have opposite effects on magnetism. Their work suggested the importance of electronic structures such as Fermi surface effects, in determining the character of the exchange coupling as well as the magnitude of magnetoresistance in the Fe / Cr superlattices.

We have, so far, reviewed the electronic and magnetic properties of thin film strucutres, especially on the Fe / Cr superlattices, overlayers, and sandwiches. There is, however, no consideration of the effects of relaxations on the fundamental electronic structures and exchange couplings in Fe / Cr systems.

In this paper, we have determined the Fe-Cr interlayer spacing for the Fe / Cr (001) system by the total-energy calculation. We have also calculated spin densities, magnetic moments, and density of states (DOS) of Fe / Cr (001) using the highly precise all-electron total-energy FLAPW method [1].

In Sec. Ⅱ we briefly describe our calculational method. The calculated results are presented and discussed in Sec. Ⅲ. A brief summary is given in Sec. Ⅳ.

2. Calculational Method

We have adopted the single slab model to investigate the surface and interface magnetism of Fe overlayer on Cr (001) which consists of 1 ML of Fe attatched to each side of five-layer Cr (001). To investigate effects of relaxation, we allowed the interlayer spacing between Fe-Cr to be relaxed, while the interlayer spacings among the Cr atoms are kept to bulk value of bcc Cr (2.72 a.u.). The total-energy minimum is determined by fitting the calculated total-energy values to a parabola.

The all-electron total-energy local-spin-density-functional equations are solved self-consistently by means of the FLAPW method for single slab geometry [1]. The core electrons are treated fully relativistically and the valence states are calculated semirelativistically [11]. Lattice harmonics with angular momentum components $l \leq 8$ are included to describe the charge and potential within the muffin-tin (MT) sphere of radius of 2.20 a.u. The wave functions are expanded in $\sim 2 \times 550$ LAPW basis functions for each of the 21 k-points in the irreducible wedge of the two dimensional Brillouin zone. We employ the explicit form of von Barth and Hedin for the local spin exchange-correlation potential and the Hedin-Lundqvist potential for the paramagnetic case [13].

3. Results and Discussions

The total-energy curve as a function of Fe-Cr interlayer spacing is given in Fig. 1. The solid circles represent our calculated data points, which are fitted to a parabola (solid line). The total-energy minimum is found at 2.23 a.u. which corresponds to 18 % downward relaxation compared to the unrelaxed Fe-Cr interlayer spacing. It is notable that such downward relaxation is much larger than that of Fe overlayer on W (110) [13] and also that of Ni / Fe (001) system [14]. In the sense of classical electromagnetism, magnetized atoms produce magnetic flux around themselves which induce mechanical pressure, known as magnetic pressure effect [15], to neighboring magnetic atoms. Since Fe and Cr layers are antiferromagnetically coupled, it is considerted that there is a stronger negative magnetic pressure which causes a larger contraction compared with the Ni / Fe (001) system [14].

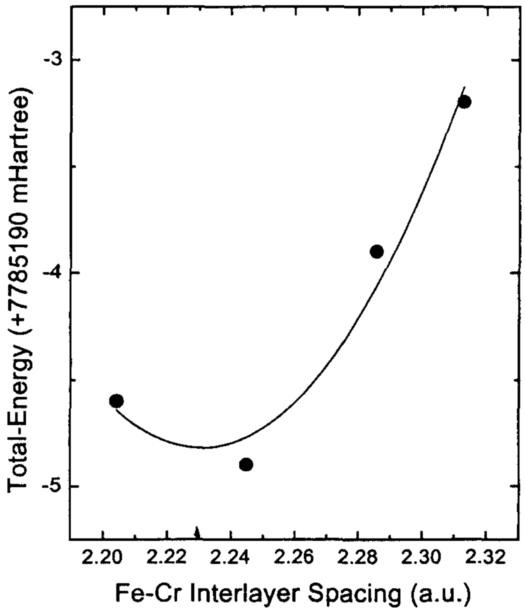
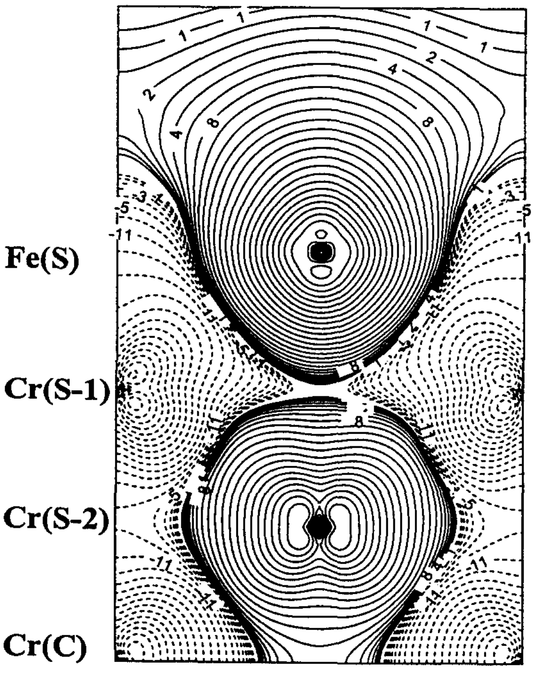
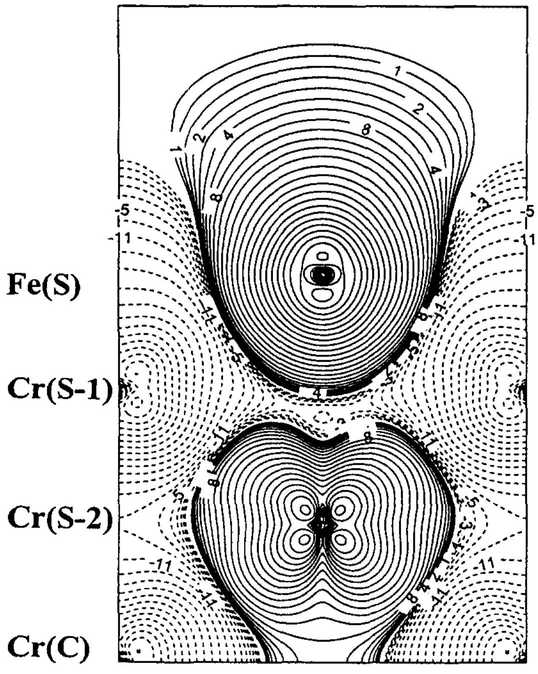


Fig. 1. Total-energy curve for the Fe / Cr (001) as a function of the Fe-Cr interlayer spacing. The solid circles represent our calculated values.



(a) Spin-Density of Unrelaxed Fe / Cr (001)



(b) Spin-Density of 17.5 % Relaxed Fe/Cr (001)

Fig. 2. Spin-density of unrelaxed (a) and 17.5 % relaxed (b) on the (110) planes in units of 1×10^{-4} electrons/a.u.^{3}. Subsequent contour lines differ by a factor of 2. The solid lines and the dashed lines represent majority- and minority-spin, respectively.

The spin density contour plots on (110) planes are presented for unrelaxed and 17.5 % downward relaxed Fe/Cr (001) in Fig. 2. There is a large extension of Fe (*S*) spin density toward vacuum region for unrelaxed system, and it becomes smaller for the relaxed system which implies a reduction of magnetic moment at Fe (*S*). We observe a negative polarization at interstitial region between Fe (*S*) and Cr (*S − 2*) for the relaxed system which causes a distortion of spin-density contours of Cr (*S − 2*). When Fe (*S*) is relaxed downward, the interaction strength among the Fe and Cr atoms is increased to distort the *d* elecron character compared with that of unrelaxed case. It is expected a small variation of magnetic moment of inner Cr layers for relaxed system.

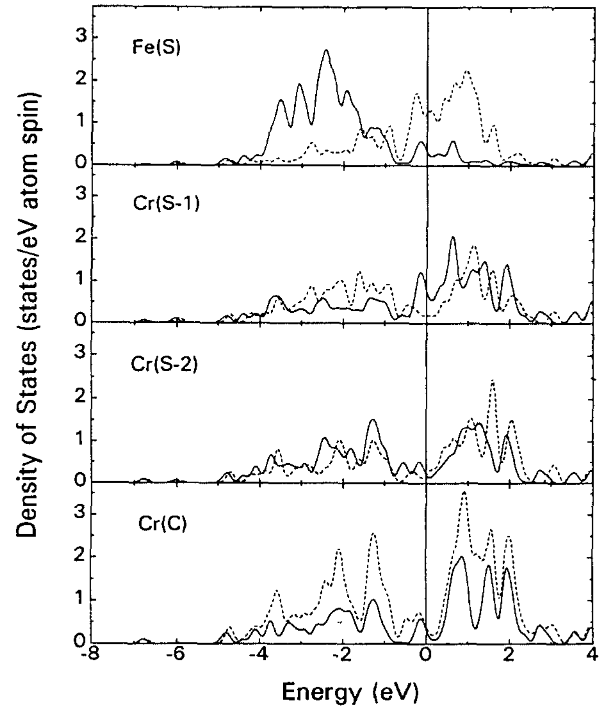
Table 1 presents the *l*-decomposed majority- and minority-spin charges inside the MT spheres, layer-by-layer magnetic moments, and work functions for unrelaxed and 17.5 % downward relaxed Fe/Cr (001). The magnetic moment of Fe (*S*) for the relaxed system is 1.72 μ_B, much suppressed from the 2.39 μ_B for the unrelaxed system. It is considered that shorter distance between the Fe and Cr layers brings about stronger hybridization between Fe-*d* and Cr-*d* states which is responsible for the reduction of surface magnetic moment at Fe (*S*) in the relaxed system.

It is found that the magnetic moment of interface Cr is slightly enhanced (0.68 μ_B) while the magnitude of magnetic moments of inner Cr layers are slightly reduced (0.58 μ_B). The work function of the relaxed system is increased by ~0.5 *e*V compared with that of the unrelaxed system.

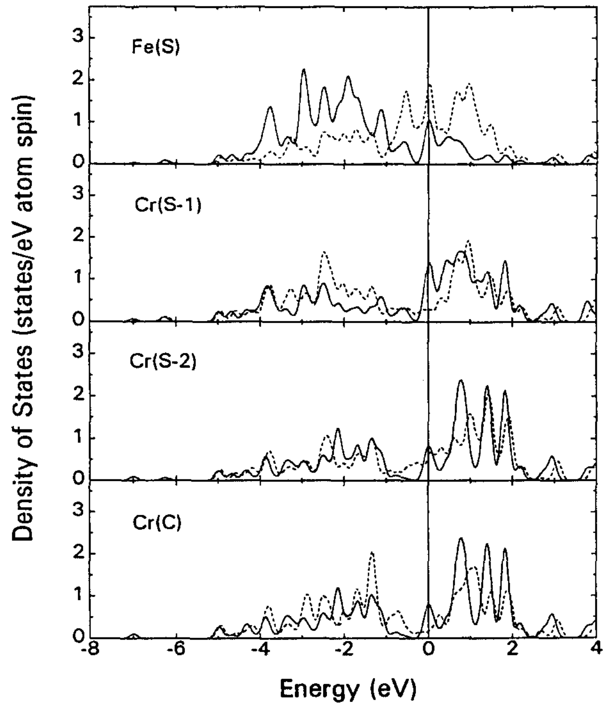
Fig. 3 shows layer projected density of states (DOS) for the unrelaxed and relaxed systems. As one may see, there is an enhanced hybridization between Fe (*S*) and Cr (*S − 1*) for the relaxed system, compared to the unrelaxed system. The DOS

Table 1. *l*-decomposed majority (↑) and minority (↓) spin charges (in electron) inside the MT sphere, layer-by-layer magnetic moment (in μ_B), and work functions (in eV) for unrelaxed and for 17.5 % downward relaxed Fe/Cr (001).

Fe/Cr (001)			*l* decomposition of charge				Magnetic moment (μ_B)	Work function (eV)
			s	*p*	*d*	Total		
unrelaxed	Fe (S)	(↑)	0.15	0.08	4.10	4.34	2.39	4.72
		(↓)	0.13	0.07	1.74	1.94		
	Cr (S − 1)	(↑)	0.11	0.11	1.55	1.78	−0.64	
		(↓)	0.12	0.12	2.17	2.42		
	Cr (S-2)	(↑)	0.13	0.12	2.19	2.45	0.65	
		(↓)	0.12	0.13	1.54	1.80		
	Cr (C)	(↑)	0.12	0.12	1.54	1.79	−0.67	
		(↓)	0.13	0.13	2.20	2.46		
17.5 % downward relaxed	Fe (S)	(↑)	0.19	0.12	3.88	4.19	1.72	5.22
		(↓)	0.17	0.12	2.17	2.47		
	Cr (S-1)	(↑)	0.15	0.15	1.62	1.94	−0.68	
		(↓)	0.16	0.17	2.28	2.62		
	Cr (S-2)	(↑)	0.15	0.16	2.24	2.56	0.58	
		(↓)	0.15	0.16	1.66	1.98		
	Cr (C)	(↑)	0.14	0.16	1.65	1.97	−0.58	
		(↓)	0.15	0.16	2.22	2.55		



(a) Unrelaxed Fe/Cr (001)



(b) 17.5 % Relaxed Fe/Cr (001)

Fig. 3. Spin-decomposed and layer-projected density of states (DOS). The majority-spin and the minority-spin are represented by solid lines and by dashed lines, respectively.

width of Fe (*S*) for the relaxed system becomes broader than that of the unrelaxed system. This enhanced hybridization is considered to be responsible for the large reduction of magnetic moment of Fe (*S*) of the relaxed system compared to the unrelaxed system.

4. Summary

The effect of surface relaxation on surface and interface magnetism in Fe/Cr (001) has been investigated using the self-consistent FLAPW method. The spin densities, magnetic moments, layer-by-layer density of states, and work functions have been calculated for both of relaxed and unrelaxed systems.

The equilibrium Fe-Cr interlayer spacing is determined by total energy calculation. It is found that the equilibrium interlayer spacing between surface Fe and subsurface Cr is 2.23 a.u. which coresonds to 18 % downward relaxation.

The magnetic moment of surface Fe for the relaxed system is calculated to be 1.72 μ_B which is much suppressed from the value of 2.39 μ_B for the unrelaxed system. It is found that the magnetic moment of interface Cr is slightly enhanced (0.68 μ_B) while the magnetic moments of inner Cr layers are slightly reduced (0.58 μ_B).

We observe strong hybridization between Fe-*d* and Cr-*d* states from the calculated DOS. This enhanced hybridization between Fe-*d* and Cr-*d* states is considered to be responsible for the large reduction of the magnetic moment of Fe (*S*) for the relaxed system.

In conclusion, our results reveal the importance of relaxation effect to the magnetism of Fe/Cr system.

Acknowledgements

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