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Crystal-field Effect on Magnetic Moment and Exchange-Coupling for Fe/W(100) and Fe/W(110)

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Abstract

The Full-potential Linearized Augmented Plane-wave (FP-LAPW) method was employed to investigate the magnetic properties of 1 monolayer (ML) Fe on W(100) and W(110) substrates. Magnetic moments of the Fe overlayer are found to be very different with ~2.0 μ_B for Fe on W(100) and ~2.56 μ_B for Fe on W(110). The exchange coupling between Fe film and W substrate are also found to be orientation-dependent. The electronic coupling in Fe/W(100) thin film is found to be more long-range, compared to the one in Fe/W(110). These differences could be explained by the differences of local atomic bonding and crystal-field splitting in these two orientations.

I. Introduction

Exchange coupling between atomic layers constitutes many interesting phenomena including Giant Magneto-resistance (GMR) and spin-waves in magnetic multilayers. It was found that both the strength and oscillation period of interlayer exchange coupling depend strongly on the thickness of the spacer layer and crystal orientation [1]. Two theories have been developed and employed to explain the interlayer exchange couplings [2,3]. One is based on the Ruderman-Kittel-Kasuya-Yosida (RKKY) spin-dependent scattering model [2]. The other is based on the quantum size effect, i.e. the modulation of density-of-states (DOS) due to quantum confinement [3]. Later Bruno [4] showed that these two theories are actually equivalent. Indeed, in electronic structure calculations, full multiple-scattering yields the density-of-states (DOS). These two theories are quite successful in explaining most of the experimental observations [4]. Despite the good agreement in GMR between theory and experiment, so far the short oscillation periods of the exchange coupling predicted by theory in the (110) oriented multilayers have not been observed in experiments [5]. Some attribute this discrepancy to the surface and interface roughness or disorder effect [6,7]. It was indeed possible to make the short oscillation period disappear when the effect of disorder was included in the calculations [6]. However, the underlying physical reason is still uncertain. Understanding exchange coupling on the short-range scale is necessary in order to understand the effects of interface atomic structure on GMR.

Besides interlayer exchange coupling, ferromagnetism in transition metals is still not completely understood [8-10]. So far the relative contribution of intra- and inter-atomic exchange interaction to magnetic moment and their effect on magnetic coupling is not entirely clear. Whether these two interactions are collaborative or competing largely depend on the system and its atomic structure under study. For a given metallic system, if inter-atomic interaction dominates, larger inter-atomic distances tend to favor ferromagnetism and an

increased magnetic moment due to a narrower *d* band. In the situation where intra-atomic interaction dominates, larger inter-atomic distance means smaller crystal-field splitting for the *d* orbitals. Therefore an enhanced magnetic moment is also expected. At a given inter-atomic spacing, the magnetic property of the system depends on many competing interactions including magnetic, electronic, elastic, and magneto-elastic interactions that eventually lead to the lowest energy state. The surface and interface effects are important when magnetism is dominated by inter-atomic exchange interaction because of its sensitivity to chemical bonding. However, interface will also affect intra-atomic interaction due to the change in crystal-field and therefore the width and splitting of the *d*-band. The temperature dependence of magnetism actually concerns largely the relative importance of inter-atomic and intra-atomic exchange interactions in magnetism [11]. The fact that Fe, Co and Ni elements in bulk or thin films possess magnetic moments less than Hund's second rule predicts for isolated atoms means that inter-atomic interaction is important in these elements.

Monolayer Fe films grown on W(100) and W(110) substrates are used as examples to illustrate the orientation dependence of the exchange coupling on a short-range scale, and to understand the effect of crystal-field on magnetic moment of the Fe overlayers.

II. The Systems

Ultrathin Fe films grown on W(100) and W(110) substrates are prototype magnetic systems for studying thin film magnetism due to their thermal stability, large lattice mismatch of 9.4%, and pseudomorphic growth. However, Fe films deposited on W(100) and W(110) substrates exhibit very different magnetic properties [12-16]. It was observed that there are magnetic 'dead-layers' in the sub-monolayer coverage of Fe on W(100) [16,17] above the experimental temperature of 115 K and 140 K respectively, while an enhancement of the surface magnetic moment was observed for the 1 ML Fe/W(110) system [13,18,19]. In both experiments [12] and theoretical calculations [18], it was found that Fe has an enhanced moment of about 2.56 μ_B in 1 ML Fe/W(110) film compared to the moment of 2.2 μ_B in bulk bcc Fe. Further, a Curie temperature of 282K was observed for the monolayer Fe on W(110) [12,13]. So far, this difference has not been extensively explored nor understood.

Since electronic structure directly reflects the atomic structure of the system, magnetism is very sensitive to the details of the atomic structure. 1 ML Fe/W(100) has a C_{4v} point-group symmetry, while 1 ML Fe/W(110) has a lower C_{2v}. Since symmetry is crucial in crystal-field splitting of the *d* orbitals, it is not unexpected that Fe/W(110) and Fe/W(100) films exhibit different magnetic properties. Indeed, according to crystal-field theory, the 3*d* orbitals will be split into different bands for different symmetry. As a result, the interaction between Fe and W will be greatly affected by the symmetry. The relative strength of the crystal-field depends on the nature of chemical bonding and geometric effects.

III. Results and Discussion

The WIEN97 code [20] based on the FP-LAPW method was used to carry out the calculations. Repeated slab geometries consisting of a total of 5 W substrate layers and 1 Fe layer on each surface were employed in all of the calculations. A total of 3000 k-points in the full Brillouin Zone was used for both Fe/W(110) and Fe/W(100) films to ensure complete convergence and a proper comparison. The Generalized Gradient Approximation (GGA) was

	$d(Fe-W_1)$ (Å)	$d(W_1-W_2)$ (Å)	$\mu_B(Fe)$	$\mu_B(W_1)$	$\mu_B(W_2)$	$\mu_B(W_3)$
Fe/W(100)	1.25	1.61	2.09	-0.25	0.090	-0.076
Fe/W(110)	1.97	2.26	2.56	-0.085	-0.000053	-0.00027

Table I. Comparison of atomic interlayer distances and magnetic moments between 1 ML Fe/W(100) and Fe/W(110).

adopted. Additional computational details were described in our earlier publications [18,21].

The theoretical equilibrium bcc W lattice constant of 3.205Å was used for the in-plane lattice parameter in 1 ML Fe/W(110) and 1ML Fe/W(100) films. The vertical lattice spacings were completely relaxed. The magnetic moments were determined by taking the majority spin numbers minus the minority spin number. Table I exhibits the interlayer atomic distances

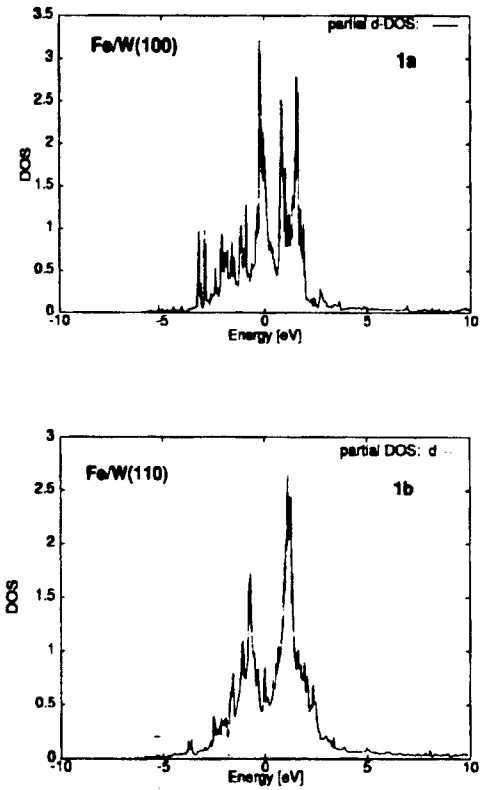

Figure 1. Fe minority-spin partial-*d* density-of-states (DOS) for 1 ML Fe/W(100) (Fig. 1a) and 1 ML Fe/W(110) (Fig. 1b)

and magnetic moments for the two systems.

As can be seen from Table I, 1 ML Fe grown on W(100) and on W(110) has very different interlayer distances and magnetic moments. In Fe/W(100) system, the Fe-W interlayer distance is much shorter compared to the one in Fe/W(110), a contraction of 36.5%. The W-W interlayer distance in Fe/W(100) is 28.8% smaller than that of Fe/W(110). It is true that the nearest-neighbor interatomic distances in both (100) and (110) oriented films are the same. However, four of the nearest-neighbor bonds lie in-plane in (110) oriented films, while all of the nearest-neighbor bonds are out-of-plane in (100) oriented films. It suggests that the interlayer interaction in Fe/W(100) is stronger compared to the one in the Fe/W(110) system because of the greater overlap between the interlayer valence orbitals in (100) oriented films than in (110) films.

Figures 1a and 1b show Fe 3d minority-spin density-of-states (DOS) for Fe/W(100) and Fe/W(110) respectively. It can be seen that the splittings of the 3*d* orbitals are different for the C_{4v} and C_{2v} symmetries as expected from crystal-field theory. There are 3 major peaks in the Fe 3*d* band grown on W(100) substrate, while only two major peaks can be observed for Fe/W(110) film. This means that crystal-field effect is important for the minority-spin DOS, which in turn largely determines the magnetic moment. Table II shows the contribution from Fe valence 4*s*, 4*p* and 3*d* orbitals to the total magnetic moment.

The Fe magnetic moments are found to be different in these two orientations with 2.09 μ_B and 2.56 μ_B for 1 ML Fe grown on W(100) and W(110) substrates respectively. The magnetic moment of Fe is enhanced by 16% in 1 ML Fe/W(110) compared to the bulk bcc Fe value of 2.2 μ_B in agreement with previous experimental results [12,13]. However, the Fe magnetic moment is much reduced for the (100) orientation. As seen from this Table II, the major contribution to the difference of Fe magnetic moments in these two films is from $3d_{xz}$ and $3d_{xy}$ orbitals. Moreover, the interfacial W atom in (110) film acquires an induced moment of 0.085 μ_B, which is much smaller compared to 0.25 μ_B for the interfacial W atoms in the (100) films. Recent experimental data [22] also demonstrate an induced interfacial W moment of ~0.2 μ_B for Fe/W(100) in good agreement with our current calculations. The inner W atoms have very small induced moments in the (110) film, while the induced moments for W persist for the inner layers in the (100) films. The second and third layer W atoms have moments of ~0.09 μ_B and ~0.07μ_B respectively in the (100) oriented films. Interestingly, the W magnetic moments are ferromagnetically coupled to each other in W(110) substrate, in contrast to the anti-ferromagnetic coupling in W(100) substrate. This difference perhaps could be understood from the different interlayer distances in these two orientations. The (100) oriented film has a much shorter inter-layer distance compared to the (110) oriented film, thus ferromagnetic coupling is favored in the (110) films when inter-atomic exchange interaction plays an important role.

The difficulty in observing the short-oscillation period of magnetic coupling in (110) oriented films may be related to the rapid-decay of induced magnetic moments in W seen here in the Fe/W(110) system. The (100) oriented film, on the other hand, sustains a substantial value of induced magnetic moment even for the inner W layers seen from Table I. The underlying reason

spin (↑-↓)	μ_B	*s*	*p*	d_{tot}	d_z^2	$d_{x^2-y^2}$	d_{xy}	d_{xz}	d_{yz}
Fe/W(100)	2.09	0.033	0.004	2.059	0.628	0.521	0.189	0.391	0.331
Fe/W(110)	2.562	0.020	-0.005	2.547	0.585	0.608	0.411	0.665	0.279

Table II. Contributions to the total Fe magnetic moment from valence *s*, *p* and *d* orbitals

could be attributed to the shorter inter-layer distances in (100) films than those in (110) films. As a result, a larger overlap of the valence-orbitals in (100) films occurs.

The magnetic 'dead-layer' observed in the submonolayer Fe coverage on W(100) substrate [16,17] could be partially attributed to the reduced moments calculated here for Fe/W(100). However, there must be other mechanisms contributing to the complete suppression of magnetic signal. It is possible that the Curie temperature for the 1 ML Fe/W(100) system is much lower than the corresponding Curie temperature of 282K for the 1 ML Fe/W(110). It is known that Curie temperature is very sensitive to the film thickness, the coordination number, the capping and substrate layer thickness, and the interatomic distance [23-27]. Since exchange coupling J decays as $\sim 1/R^3$ [28], in which R is the interatomic distance, the Curie temperature will be dependent upon the crystallographic orientation of the thin film as demonstrated in [27]. If only nearest-neighbor coordination and distance are considered, then the Curie temperature for 1 ML Fe/W(100) will be around or below 100K, which is below the previous experimental temperature [16,17]. The lower magnetic moment for the monolayer Fe on W(100) substrate will reduce the Curie temperature even further compared to the corresponding W(110) substrate case [27]. All in all, our theoretical calculation does not find a magnetically dead monolayer Fe on W(100). However, it is possible that the Curie temperature of this system is far below the 1 ML Fe/W(110) case and below the experimental temperature conducted.

IV. Summary

It is shown that crystal-field effect is important for the magnetic moments and interlayer exchange coupling in ultrathin Fe/W(110) and Fe/W(100) magnetic films. The crystal-field energy splitting of the outer d orbitals and their relative positions are strongly symmetry dependent. As a result, the magnetic interactions including exchange coupling and magnetic moments are strongly affected by the crystal-field splitting and therefore by the symmetry of the thin films.

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