Exchange Interactions in the Bcc Fe/TaW(001) System*

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In a recent study Ferriani et al. (Phys. Rev. Lett. 99 (2007) 187203) investigated the possibility of tuning the magnetic order of the Fe monolayer on the disordered bcc-Ta

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those magnetic configurations in which the two Fe atoms sume that the Eq. (2) is conditionally averaged only over local magnetic moment on the Fe atom. We therefore aside of Eq. (2) would be zero in the DLM state if the averaged Green function \( \bar{G}_{\mu\nu}(\epsilon) \) method in terms of the potential functions (SGF) method \([7]\) which employs a realistic semiinfinite sample geometry. The one-electron potential is treated within the atomic sphere approximation, but the dipole barrier due to the redistribution of the electron density in the surface region is included. The TB-LMTO-SGF method can include the effect of layer relaxations approximately provided that they are known either from the full-potential calculations or from the experiment.

An important advantage of the TB-LMTO-SGF approach is the possibility to calculate exchange interactions between magnetic atoms in the overlayer. We describe these interactions in terms of the two-dimensional classical Heisenberg Hamiltonian,

\[
H = -\sum_{i\neq j} J_{ij} \mathbf{e}_i \cdot \mathbf{e}_j ,
\]

where \( J_{ij} \) denotes the exchange integral between Fe spins at sites \( i \) and \( j \) on the surface, and \( \mathbf{e}_i \) and \( \mathbf{e}_j \) are unit vectors in the directions of the local magnetization on sites \( i \) and \( j \), respectively.

The exchange interactions \( J_{ij} \) are evaluated in the TB-LMTO-SGF method as

\[
J_{ij} = \frac{1}{4\pi} \text{Im} \int_C \text{tr}_L \Delta_i(z) \tilde{g}_{ij}^1(z) \Delta_j(z) \tilde{g}_{ji}^1(z) \, dz ,
\]

where \( \Delta_i(z) \) represents the exchange splitting of the Fe atom at the site \( i \), \( \tilde{g}_{ij}^1(z) \) is the configurationally averaged Green function describing the motion of an electron between sites \( i \) and \( j \) in the magnetic overlayer on a random nonmagnetic substrate which corresponds to the spin \( \sigma = (\uparrow, \downarrow) \), and the integration is done over the contour \( C \) in the complex energy plane \( z \) which starts below the valence band and ends at the Fermi energy. Symbol \( \text{tr}_L \) denotes the trace over the atomic orbitals \( (L \equiv (l, m)) \). The quantity \( \Delta_i(z) \) is defined in the TB-LMTO method in terms of the potential functions \( P_{i\alpha}^m(z) \) as \( \Delta_i(z) = P_{i\uparrow}^m(z) - P_{i\downarrow}^m(z) \). The reader is referred to Refs. 7, 8 for details of evaluation of the real-space configurationally averaged Green function \( \tilde{g}_{ij}^1(z) \) for the magnetic overlayer on a random substrate. The expression on the right-hand side of Eq. (2) would be zero in the DLM state if the averaging were performed over all possible orientations of the local magnetic moment on the Fe atom. We therefore assume that the Eq. (2) is conditionally averaged only over those magnetic configurations in which the two Fe atoms have parallel magnetic moments.

III. RESULTS AND DISCUSSION

The calculations were done assuming the Vegard’s law for the bulk lattice constant \( a \) of bcc-Ta\(_x\)W\(_{1-x}\) random alloy \( (a_{Ta} = 3.300 \text{ Å} \text{ and } a_{W} = 3.165 \text{ Å} ) \). In all calculations we have employed the \textit{spdf}-basis and the Vosko-Wilk-Nusair exchange-correlation LSDA potential. Similarly as in Ref. 2, we assume a constant inward layer relaxation of 18% between the Fe-overlayer and the bcc(001)-Ta\(_x\)W\(_{1-x}\) substrate.

The total DOS of bulk W and Ta atoms are rather similar, which shows that the virtual crystal approximation used in the Ref. 2 is well justified. The calculated local densities of states (LDOS) show that the hybridization between the magnetic Fe monolayer and W substrate is stronger than the hybridization with the Ta substrate. The main difference between the W and Ta substrates is the position of the Fermi energy which is shifted downwards in the bcc-Ta because of smaller number of valence electrons. As a consequence, the Fermi energy lies inside the bonding peak of the bcc Ta while it is shifted to the energy region between the bonding and antibonding states in the bcc W.

A. Exchange integrals

Concentration dependence of the exchange integrals \( J_s \) for the first 5 shells is shown in Fig. 1. The exchange integrals are not limited to the first two shells as in [2]. In the two-dimensional case the exchange integrals can decay more slowly than in the three dimensional case (see e.g. Ref. 9).

![FIG. 1: Exchange integrals between Fe-atoms (up to the 5th shell) in the Fe overlayer on the Ta\(_x\)W\(_{1-x}\) (001) random alloy surface as a function of the substrate alloy composition.](http://www.sssj.org/efdssnt)
the crossover between the FM and AFM ground state of the Fe overlayer. The concentration dependence of other interactions on the substrate alloy composition is rather weak. The second NN interaction is of the AFM character in the whole concentration range while the third NN interaction is FM-like for a W-rich substrate. The first two NN interactions agree reasonably well with those fitted from total energies for three magnetic configurations [2] (note that the definitions of the Heisenberg Hamiltonian used in the present paper and in Ref. 2 differ by a factor of 2).

B. Magnetic stability

The lattice Fourier transform of the site-dependent exchange integrals

$$J(q) = \sum_{j} J_{0j} e^{i q \cdot R_{0j}}$$

(3)
is closely related to the magnon spectrum of the system described by the HH, Eq. (1). The maximum of $J(q)$, or, equivalently, the minimum of $-J(q)$, reached for a particular value of the vector $q$ in the surface Brillouin zone (SBZ) indicates a tendency of the magnetic system to form a magnetic ground state characterized by the wave vector $q$. The wave vector $q = 0$ (point $\bar{\Gamma}$ in the SBZ) corresponds to the FM ground state while a nonzero wave vector $q$ corresponds to a more complex ground state including possible AFM or spin-spiral states. For example, $q$ at the point $M$ of the SBZ corresponds to the $c(2 \times 2)$ AFM ground state (checkerboard, i.e., each atom has 4 nearest neighbors with opposite moments, see Fig. 2b in Ref. 10) and $q$ at the point $X$ corresponds to the $p(2 \times 1)$ AFM ground state (row-wise, i.e., atoms in odd rows have magnetic moments up, in even rows have moments down, see Fig 2c in Ref. 10). The minimum of $-J(q)$ located in a general point of the SBZ indicates an incommensurate (spin-spiral) magnetic ground state.

In this way, we can investigate the stability of a much broader class of magnetic configurations than by the conventional total energy search. We mention that the relativistic effects, for example, strengthen the tendency towards collinear AFM alignment and compete with the DMI interaction which promotes the rotating, non-collinear magnetism as it was demonstrated recently for Fe double chains on the fcc-Ir(001) surface [13]. If the Ta content further increases, the $p(2 \times 1)$-AFM configuration becomes the ground state as it is illustrated for the case with 50% of Ta atoms where the minimum of $-J_{Fe,Fe}(q)$ occurs at $X$, see Fig. 2. In the Ta-rich alloy the stability of the FM phase increases as compared to the $c(2 \times 2)$ configuration until for Fe/Ta(001) the FM state becomes the magnetic ground state.

The observed tendency to a non-collinear ground state for $x \approx 0.2$ can be explained by frustration effects on the square Fe lattice accompanying the dominating first and second nearest-neighbor interactions that are both negative for concentrations $x \leq 0.45$, see Fig. 1. For W-rich substrates ($x \to 0$), the first nearest-neighbor interaction dominates ($|J_1| > |J_2|$) and the $c(2 \times 2)$ AFM ground state is not frustrated. For equiatomic concentrations ($x \approx 0.5$), the second nearest-neighbor interaction is the strongest one ($|J_2| > |J_1|$) and the $p(2 \times 1)$ AFM ground state is not frustrated either. However, for compositions around $x \approx 0.2$, both interactions are of comparable magnitude ($J_1 \approx 2J_2$) which leads to a strong frustration of these simple AFM states.

This analysis is further supported by the phase diagram of the Ising Hamiltonian with exchange interactions between the first and second nearest neighbors (see, e.g., the phase diagram at $T = 0$ in variables $J_1$ and $J_2$ of Ising model, Fig. 3 in Ref. 10). Note that the Heisenberg model is equivalent to the Ising model, if we consider only the collinear phases. If $J_1 \approx 2J_2$ and $J_1 < 0$, $J_2 < 0$, the system is close to the transition between the $c(2 \times 2)$ and $p(2 \times 1)$ states. In this situation even small higher-order exchange parameters $J_n$, $n > 3$ can play a role and give rise to a more complex magnetic structure.
IV. CONCLUSIONS

We have studied magnetic phase stability of magnetic overlayers on non-magnetic disordered substrates. The approach consists in the evaluation of exchange integrals between local magnetic moments in the magnetic overlayer using the adiabatic approximation and the real-space Green function approach. The exchange interactions between pairs of local moments in the overlayer were used to construct the effective two-dimensional Heisenberg Hamiltonian, whose stability with respect to periodic spin excitations was investigated. The maxima of the lattice Fourier transform of exchange integrals served as indications of stable periodic spin structures. This approach allowed us to investigate the stability of a much broader class of magnetic configurations than the conventional total energy search limited to a few configurations. As a case study we have investigated in detail the magnetic phase stability of the Fe overlayer on the bcc-Ta$_x$W$_{1-x}$(001) random substrate. The exchange interactions for this system were extracted from the disordered local moment state. Our results are in a good agreement with a recent study [2] based on the total energy search. In addition, we have predicted a possible incommensurate magnetic configuration for the W-rich substrate alloy (at about 20% of Ta).

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