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First-principles Calculations of the Exchange Coupling Constants for an Iron Film

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Abstract. First-principles calculations of the energy and magnetic characteristics of Fe films on Cr substrates are carried out with the use of the VASP software package in PAW PBE approximation. The exchange interaction parameters for Fe/Cr(001) and Fe/Cr(001)/Fe structures as a function of ferromagnetic films thickness were calculated within a classical Heisenberg model. The results of our calculation of exchange coupling constants can be used to calculate the temperature dependence of magnetoresistance and compare them with experimental data.

Keywords: ultrathin films, ab-initio calculations, exchange interaction parameters, magnetoresistance.

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Introduction

Today, ultrathin magnetic films are the object of intensive research, both in Russia and abroad. The results obtained by researchers in this area are discussed at Russian and international conferences, numerous reviews, articles and monographs are written. Such great interest is due to the fact that the study of the properties of ferromagnetic films contributes to the solution of fundamental problems in the physics of magnetic phenomena and the development of the theory of ferromagnetism. The study of films provides new information about the magnetic properties of ferromagnets, deepening our knowledge in the field of magnetism. Thus, the study of thin films has significantly expanded the understanding of the physical nature of the anisotropy of ferromagnets, made it possible to identify and study magnetization reversal processes and discover new physical phenomena. One of these phenomena is giant magnetoresistance (GMR), which occurs in structures consisting of ferromagnetic layers separated by non-magnetic layers. Such magnetic structures are widely used as read heads of hard disks, memories, and spintronics devices. In this work, the atomic structures and magnetic properties of iron films on a Cr substrate are studied to describe exchange-correlation interactions.

A very important parameter in magnetism, in particular in micromagnetism, is the exchange interaction constant. This parameter reflects the strength of the connection between neighboring spins, which arises due to exchange interaction. The exchange interaction constant is a phenomenological parameter reflecting the magnetic symmetry of the system. It can be related to the microscopic parameters of the system, for example, to the exchange integral in the Heisenberg model. Today's interest in thin magnetic films and nanostructures makes it important to

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understand how the exchange interaction constant behaves as the size of the system changes and in the presence of interfaces.

It is very difficult to obtain the exchange interaction constant experimentally. There are very few works in which the constant for thin magnetic films [1] is estimated. Theoretically, as a rule, the exchange interaction constant is estimated by indirect methods, which include assumptions about specific models of magnetic order. There are various software packages for obtaining energy values and finding exchange interactions. For example, in the work [2] a study of exchange interaction was carried out using the Corring–Kohn–Rostoker (KKR) method. In this paper, the authors calculated the Heisenberg exchange parameter for bcc-Fe, fcc-Co, and fcc-Ni. In the work [3], using density functional theory (DFT) with the full-potential linearized augmented-plane-wave (FLAPW) method, the exchange interaction was calculated for bulk bcc-Fe and Fe/Au. It should be noted that DFT gives slightly overestimated results in relation to the mean field method in comparison with the KKR method, which describes volumetric structures well. However, for calculating thin-film structures, the KKR method works worse, so in this work we performed calculations by the VASP (Vienna Ab-Initio Simulation Package) software package [4] using the Projector AugmentedWave (PAW) method with the PBE version generalized gradient approximation (GGA).

1. Model

In this work, within the framework of a first-principles approach, using the spin density functional method, the formation of ultrathin ferromagnetic films on the surface of a non-magnetic substrate was theoretically described. The occurrence of exchange interaction in bulk material, as well as in films, has been studied.

In this work, the following structures were studied: a bulk 16-atomic bcc cell of iron, as well as a film of iron, both pure and adsorbed on a chromium surface with a (100) face orientation. Fig. 1 shows the crystal structure of the supercells of the Fe/Cr and $Fe/Cr/Fe$ systems.

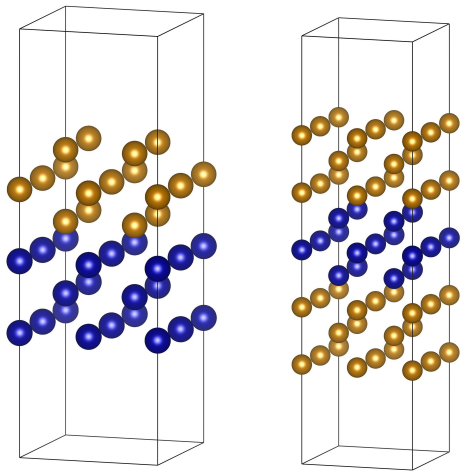

Fig. 1. The crystal structure of a supercell

Calculations were carried out for a chromium substrate with a thickness of 3 monolayers, while the thickness of the iron film varied from one to three monolayers. There is also a 5Åvacuum layer on both sides. Also on both sides there is a vacuum layer of 5 Å. We obtained the value of the lattice constant a=5.74 Å.

Integration in the first Brillouin zone was carried out using a k-grid constructed using the Monkhorst–Pack method. The k-grid size was $13 \times 13 \times 1$. The plane-wave basis cutoff energy was chosen to be 500 eV.

The contribution to ab initio total energies, corresponding to exchange interaction, can be described by the following classical Heisenberg model:

$$H = -\sum_{i,j} J_{ij}(\mathbf{S}_i \mathbf{S}_j). \tag{1}$$

We got a system of equations for determination of exchange interaction integrals by calculating the difference in energies of the ferromagnetic (FM) and antiferromagnetic (AFM) configurations of spins in the film:

$$J_j = \Delta N_{ji}^{-1} \Delta E_i, \tag{2}$$

where

$$\Delta N = N_{AFM} - N_{FM}, \tag{3}$$
$$\Delta E = E_{AFM} - E_{FM}, \tag{4}$$

N is calculated as

$$N = -\sum_{ij} S_i S_j. \tag{5}$$

For the case of taking into account the interaction of the nearest neighbors ($j = 1$) and the next nearest neighbors ($j = 2$) we obtain the following expression:

$$\begin{pmatrix} J_1 \\ J_2 \end{pmatrix} = \begin{pmatrix} \Delta N_1 & \Delta N N_1 \\ \Delta N_2 & \Delta N N_2 \end{pmatrix}^{-1} \begin{pmatrix} \Delta E_1 \\ \Delta E_2 \end{pmatrix}. \tag{6}$$

The antiferromagnetic spin configuration for which the calculation was carried out is shown in Fig. 2. We will describe in more detail the mechanism of calculating the number of nearest

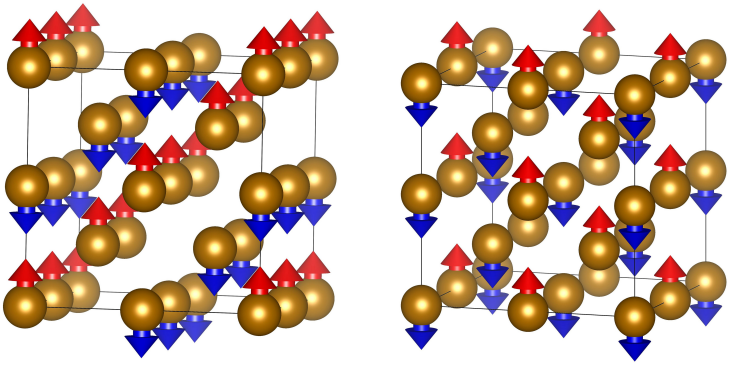

Fig. 2. Orientations of magnetic moments of atoms

neighbors. We gradually increased the radius of the coordination sphere until the array of neighbors increased, then we checked which new atoms were included in the array of neighbors. If the new atoms corresponded to the selection, then the iterations were stopped, if the increase was due to atoms that should not be taken into account in the calculations, then they were removed from the array and the search continued. For example, consider the 4Fe12Cr system. We need to get Fe-Fe neighbors. Iron atoms have indices 0, 1, 2 and 3. Gradually increasing the

radius of the coordination sphere, we get a list of neighbors for each atom. The mechanism is clearly described in (7).

$$\begin{bmatrix} 1 & 2 & 4 & 8 & \cdots \\ 3 & 4 & 2 & 9 & \cdots \\ 3 & 4 & 0 & 10 & \cdots \\ 4 & 5 & 6 & 7 & \cdots \\ 5 & 2 & 8 & 9 & \cdots \\ 7 & 9 & 11 & 13 & \cdots \\ 7 & 10 & 11 & 14 & \cdots \\ 11 & 15 & 1 & 5 & \cdots \\ 9 & 10 & 12 & 0 & \cdots \\ 11 & 12 & 13 & 8 & \cdots \\ 11 & 12 & 13 & 8 & \cdots \\ 12 & 13 & 14 & 15 & \cdots \\ 13 & 14 & 4 & 8 & \cdots \end{bmatrix} \Rightarrow \begin{bmatrix} 1 & 2 & \_ & 0 \\ 3 & \_ & 2 & \_ \\ 3 & \_ & 0 & \_ \\ \_ & \_ & \_ & 1 \\ \_ & 2 & \_ & \_ \\ \_ & \_ & \_ & \_ \\ \_ & \_ & \_ & \_ \\ \_ & \_ & 1 & \_ \\ \_ & \_ & \_ & 0 \\ \_ & \_ & \_ & \_ \\ \_ & \_ & \_ & \_ \\ \_ & \_ & \_ & \_ \\ \_ & \_ & \_ & \_ \end{bmatrix} \Rightarrow \begin{bmatrix} 1 & 2 & \_ & 0 \\ 3 & \_ & 2 & \_ \\ 3 & \_ & 0 & \_ \\ \_ & \_ & \_ & 1 \end{bmatrix} \quad (7)$$

Each i-th row of the array is a set of indices of atoms that are neighbors of one i-th atom. After receiving the array, we remove all chromium atoms that are neighbors, that is, in each row we remove elements that correspond to the indices of chromium atoms. This leaves us with an array of Fe-Fe and Cr-Fe neighbors. After that, we delete the chromium atoms themselves, that is, the rows whose indices correspond to the indices of chromium atoms. As a result, we have Fe-Fe neighbors.

Next, we sort through the remaining pairs of atoms, if the spin direction is co-directed, then we take –1, if it is oppositely directed, then 1.

The values of these sums for the systems under study are given in Tab. 1.

Table 1. The number of pairs of the Fe-Fe nearest (NN) and next- nearest neighbors (NNN) with oppositely directed spins

		1 ml		2 ml		3 ml	
		NN	NNN	NN	NNN	NN	NNN
Fe	FM	–16	–16	–32	–64	–64	–136
	AFM_1	16	–16	0	0	0	24
	AFM_2	0	16	–16	48	–32	88
FeCr	FM	–16	–16	–32	–64	–64	–136
	AFM_1	16	–16	0	0	0	24
	AFM_2	0	16	–16	48	–16	72
FeCrFe	FM	–32	–32	–64	–128	–128	–272
	AFM_1	32	–32	0	0	0	48
	AFM_2	0	32	–32	96	–32	144

Thus, subtracting the values N for AFM and FM configuration, we get ΔN.

2. Result

Using the methodology described above, we calculated and presented in Tab. 2 the values of of the exchange integral J_1 for interaction of the nearest neighbours and J_2 for interaction of the next-nearest neighbours for bulk iron.

Table 2. Values of the exchange integral for the nearest neighbors J_1 and for the next nearest neighbors J_2 for Fe in the bulk

	J1			J2		
	meV	erg$*10^{-14}$	mRy	meV	erg$*10^{-14}$	mRy
Fe_{bulk}	25.117	4.023	1.846(VASP) 0.862(MF) [1]; 1.23(KKR) 1.43(KKR) [2] 4.209(FLAPW) [3];	–3.163	–0.506	–0.232

Comparison of the results of our calculations J_1 for bulk iron using VASP and KKR methods with the results of calculations by other authors showed that the values differ depending on the method used. The results obtained by the KKR method are closer to the results obtained by the mean field approximation (MF). The results obtained using VASP, in turn, slightly overestimate the values. But this overestimation is much less than the values also obtained within the framework of the DFT FLAPW method [3].

Then we obtain the values of the exchange integral J_1 and J_2 for the iron film, both pure and adsorbed on a chromium surface with a (100) face orientation for the Fe/Cr and Fe/Cr/Fe systems Tab. 3.

Table 3. Values of the exchange integral for the nearest neighbors J_1 and for the next nearest neighbors J_2 for Fe films depending on the thickness

		J1			J2		
		meV	erg$*10^{-14}$	mRy	meV	erg$*10^{-14}$	mRy
Fe	1 ml	31.343	5.021	2.303	–0.928	-0.148	–0.0682
	2 ml	43.185	6.918	3,174	–1.904	–0.3051	–1.399
	4 ml	42.410	6.794	3.117	–9.981	–1.599	–0.733
FeCr	1 ml	–9.221	–1.477	–0.677	–2.319	–0.371	–0.170
	2 ml	13.626	2.182	1.001	5.739	0.919	0.421
	3 ml	30.789	4.932	2.263	6.541	1.048	0.480
FeCrFe	1 ml	–10.399	–1.665	–0.764	–0.614	–0.098	–0.045
	2 ml	20.390	3.266	0.149	5.540	0.887	0.407
	3 ml	17.981	2.881	0.132	5.184	0.830	0.381

Tab. 4 shows the energy difference values per atom for the Fe/Cr and $Fe/Cr/Fe$ systems. Comparing the obtained results J_1 and ΔE we see that the value of ΔE directly affects the sign of the obtained values J_1. For example, in a monolayer film, for which ΔE turned out to be negative, that is, the AFM state turned out to be more energetically favorable, the value of J_1 also turned out to be negative.

Consider the values of the magnetic moments of the systems under study, given in the Tab. 5. It can be seen that the values of the magnetic moments in the outer layer of the film are almost the same as the values of the magnetic moments in the pure film, and the iron layers located closer to the substrate have a lower magnetic moment due to the magnetization of the substrate atoms.

Table 4. The difference between the reduced energies of the AFM and FM configurations for the Fe/Cr/Fe and Fe/Cr systems depending on the thickness of the Fe film. The obtained values of the radius of the coordination sphere r for the nearest and next-to-nearest neighbors

	FeCrFe. ΔE/At, eV	FeCr. ΔE/At, eV	r_1, Å	r_2, Å
1 ml	–0.133	–0.073	1.2	2.4
	–0.074	–0.055		
2 ml	0.287	0.160	0.9	1.8
	0.270	0.172		
3 ml	0.440	0.502	0.9	1.8
	0.431	0.473		

Table 5. Values of magnetic moments of atoms for different configurations depending on the thickness of the iron film

		Fe			$2Fe$				$3Fe$				
		μ_{Fe}	μ_{Cr1}	μ_{Cr2}	μ_{1Fe}	μ_{2Fe}	μ_{Cr1}	μ_{Cr2}	μ_{1Fe}	μ_{2Fe}	μ_{3Fe}	μ_{Cr1}	μ_{Cr2}
Fe	FM	3.177			2.804	2.804			2.951	2.306	2.951		
	AFM_1	3.191			2.522	2.522			2.744	1.498	2.744		
	AFM_2	3.178			2.486	2.486			2.889	1.755	2.889		
FeCr	FM	1.815	–0.646	0.150	2.903	1.781	–0.423	0.411	2.917	2.359	2.204	–1.119	1.381
	AFM_1	2.294	0.000	–0.055	2.866	1.254	–0.707	0.089	2.788	1.470	1.703	–0.040	0.226
	AFM_2	2.056	0.000	–0.009	2.903	1.543	–0.116	0.006	2.735	1.872	1.781	–0.173	0.033
FeCrFe	FM	2.258	–0.869	0.754	2.877	1.948	–0.553	0.327	2.923	2.370	2.182	–0.992	0.817
	AFM_1	2.498	0.000	0.000	2.874	1.276	–0.056	0.108	2.796	1.507	1.955	–0.053	0.217
	AFM_2	2.538	0.000	0.000	2.828	1.624	–0.205	0.086	2.744	1.862	1.866	–0.014	0.000

Due to the fact that chromium is a layer-by-layer antiferromagnet, there should be an alternation of the signs of the magnetic moments in different layers, which we see in the Tab. 5. The outer and middle layers of the chromium substrate have opposite signs.

When studying a monolayer film, we found that in antiferromagnetic cases, chromium layers have a zero magnetic moment and no magnetization occurs, which is probably why the AFM state is energetically more favorable for a monolayer film.

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Первопринципные расчеты констант обменного взаимодействия для пленки железа

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Аннотация. Используя программный пакет VASP в приближении PAW PBE, были проведены первопринципные расчеты энергетических и магнитных характеристик пленок Fe на подложках Cr. В рамках классической модели Гейзенберга рассчитаны параметры обменного взаимодействия для структур Fe/Cr(001) и Fe/Cr(001)/Fe в зависимости от толщины ферромагнитных пленок. Результаты расчета констант обменного взаимодействия могут быть использованы для расчета температурной зависимости магнитосопротивления и сравнения их с экспериментальными данными.

Ключевые слова: ультратонкие пленки, ab-initio расчеты, параметры обменного взаимодействия, магнитосопротивление.