First-principles studies of the interlayer exchange coupling in fine-layered Fe/Au multilayers

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Using the *ab initio* approach, we calculate the bilinear coupling constant for the Fe_n/Au_m multilayers with $n=2$, 4, and 6, and m ranging from 2 up to 11 monolayers. The calculations show that the ferromagnetic order of two adjacent Fe layers is energetically favored in systems with small Au thickness, up to 5 or 6 monolayers. The calculated results also very closely reproduce the strength of the magnetic couplings known from experimental data. Simultaneously, the magnetic profile of each multilayer is calculated, showing enhanced Fe magnetic moments close to the interface in comparison with the bulk values and very slight polarization of Au layers.

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I. INTRODUCTION

Artificial multilayered structures consisting of magnetic layers separated by nonmagnetic metal spacers have attracted much interest due to their unique physical properties.[1,2] Among them, the Fe/Au multilayer structures have become the subject of very intense research. The special interest in the Fe/Au structures arises from the fact that the atomic positions in the (001) planes of the bcc-Fe crystal and the fcc-Au crystal differ only by about 0.6%. Thus there is practically no additional stress induced by small mismatches between atomic positions on the growth process and properties of Fe/Au epitaxial structures. Thus very extensive studies, both experimental and theoretical, of oscillating exchange coupling,[3–6] spin dependent electron transport,[7,8] and the large magneto-optical anisotropy[9,10] have been performed for those structures.

Experimental studies of magnetic interface coupling are performed using Fe_n/Au_m superlattices,[11] or more often using simpler trilayer structures.[3,12] The experiment performed using the trilayer sample, with the thickness of the middle Au layer varied over 80 monolayers, showed 60 changes of the coupling sign.[12] The analysis of this data leads to the conclusion that two oscillatory contributions are observed. First, more pronounced short-period oscillations with $\lambda_s=2.48$ Au monolayers and second, long-period oscillations with $\lambda_l=8.6$ Au monolayers are presented.[12]

The interlayer exchange coupling is believed to be of indirect exchange interaction type and mediated by the conduction electrons of the spacer layer. The theoretical efforts relied mostly on model calculations based on the Ruderman-Kittel-Kasuya-Yosida (RKKY) method extended to multilayered systems.[13,14] A detailed analysis for large spacer thicknesses and semi-infinitive thicknesses of magnetic layers (the asymptotic limit) shows that the oscillations are directly related to the geometry of the Fermi surface of the spacer materials.[13] The calculated oscillation periods are in very good agreement with experiments, but, the theoretical estimates of the coupling strengths differ remarkably from the experimental values.[15]

Since it is generally accepted that, in the asymptotic limit, the experimentally observed oscillatory behavior of the coupling is governed by the spacer Fermi surface, there still remain questions concerning the limit of small spacer thicknesses which are comparable or shorter than real-space distances corresponding to the relevant distances of the Fermi surface. The experiments performed with Fe_n/Au_n $(n=1,5)$ superlattices showed that the interlayer coupling exhibits the oscillatory behavior, although the coupling is always ferromagnetic for $n<5$.[11]

The multilayers with a small repetition period, in which individual layers consist only of a few atomic layers, can be modeled theoretically using *ab initio* local spin density approximation calculations. However, the exact estimation of the interlayer exchange coupling, being the difference between total energies of two magnetic configurations, requires very precise calculations of the total energy of each magnetic arrangement with an accuracy below 0.1 meV. Here, we present the results of the total energy calculations performed for the Fe_2/Au_m, Fe_4/Au_m, and Fe_6/Au_m $(m=2-11)$ multilayers with both parallel and antiparallel magnetic moment alignment of two adjacent Fe layers. We find that the ferromagnetic state is energetically favored for small spacer thicknesses, up to 5 monolayers. For 6 monolayers of Au spacers, multilayers with thin magnetic layers of 2 or 4 Fe monolayers change their magnetic arrangement to antiferromagnetic, while thicker multilayers of 6 Fe monolayers stay ferromagnetically ordered. Next, we calculate the strength of the bilinear coupling constant which is in good agreement with the existing experimental data. The calculations also allowed us to determine the profiles of the magnetic moments.

II. COMPUTATIONAL DETAILS

The calculations were performed on the tetragonal bct-like supercells consisting of $n=2$, 4, or 6 Fe monolayers, and m Au monolayers ranging from 2 to 11, 10, or 8, respectively, stacked alternately along the z-direction. The Fe atoms of each Fe layer were assumed to be ferromagnetically ordered. The magnetic unit cells were doubled in the z-direction, since the arrangement of the magnetic moments of two neighboring Fe layers could be parallel or antiparallel. These unit cells were repeated periodically since the three-dimensional boundary conditions were imposed. The starting value of the in-plane lattice constant was set to 2.87 Å. The distance between two neighboring Au layers was initially set

TABLE I. The difference of total energies, $\Delta E = E_{AF} - E_F$, between the antiferromagnetic and ferromagnetic configuration of the magnetic moments of two iron layers separated by a Au spacer of increasing size. In paranthesis, the number of atoms forming each supercell is presented.

	ΔE (meV/supercell)			
m	2	4	6	8
Fe_2/Au_m	18.33 (8)	11.87 (12)	−4.69 (16)	−1.22 (20)
Fe_4/Au_m	19.47 (12)	11.08 (16)	−2.90 (20)	−6.72 (24)
Fe_6/Au_m	19.89 (16)	3.47 (20)	6.57 (24)	−4.60 (28)

to 2.04 Å (the value corresponding to the interlayer distance in an fcc-Au crystal), the distance between two neighboring Fe layers was 1.43 Å (the interlayer distance in a bcc-Fe crystal), and the Au-Fe interlayer spacing was 1.73 Å.

The optimizations of considered supercells were performed using the VASP package[16,17] within the generalized gradient approximation method of the total energy calculations. We included 11 valence electrons for Au atoms (s^1d^{10}) and eight for Fe atoms (d^7s^1) represented by plane wave expansions with an energy cutoff of 350 eV. The wave functions in the core region were obtained by the full-potential projector augmented wave (PAW) method.[18] The Brillouin zone was sampled by $12\times12\times4$ and $12\times12\times2$ Monkhorst-Pack meshes for structures with a c-axis shorter and longer than 20 monolayers, respectively. During the optimization, the Helmann-Feynman forces and the stress tensor were calculated. The crystal structure optimization was finished when residual forces were less than 10^{-2} eV/Å and the stress was less than 0.9 kb.

III. RESULTS

A. Interlayer exchange coupling

We started the calculations from the optimization of Fe_n/Au_m supercells, either without magnetic ordering, with ferromagnetic ordering, or with antiferromagnetic ordering of adjacent Fe layers. The relaxed structures are characterized by the in-plane lattice parameters which were varied from 2.90 to 3.02 Å. The distances between two neighboring layers were found to be $d_{\text{Fe-Fe}}=1.3-1.4$ Å, $d_{\text{Au-Au}}=2.02-2.10$ Å, and $d_{\text{Fe-Au}}=1.72-1.73$ Å, respectively. These structural data are in accordance with the values obtained previously for bulk fcc-Au and bcc-Fe crystals, and for Fe/Au multilayered structures with spacer thicknesses up to 4 monolayers.[19]

We found that the total energy for nonmagnetic structures is always higher, by about 0.5 eV per supercell, than the energy of the other two magnetic configurations. The difference between total energies of ferromagnetic and antiferromagnetic arrangements is more subtle. Its value ranges from 1 up to 20 meV per supercell (Table I). The ferromagnetic ordering is predicted in multilayers with small Au spacer thicknesses, up to 5 monolayers. For 6 Au monolayers, a distinct magnetic ordering is observed for structures with 2 or 4 Fe monolayers and 6 Fe monolayers. The Fe_6/Au_6 is still ferromagnetic, while Fe_2/Au_6 and Fe_4/Au_6 are antiferromagnetically ordered. Then for 8 Au monolayers, the antiferromagnetic arrangement is the lowest energy configuration.

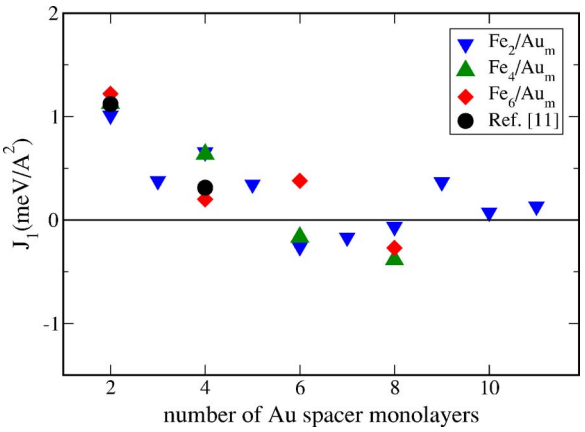

FIG. 1. (Color online) The bilinear coupling constant J_1 in Fe_n/Au_m multilayers as a function of thickness of the Au spacer layer. Solid circles denote the experimental data (Ref. 11), triangles and diamonds represent results of our calculations.

The calculations for supercells longer than 20 monolayers are very time-consuming because of the increasing number of atoms. Moreover, the convergence of the electronic structure becomes very slow. Thus we did not continue the calculations for supercells longer than Fe_2/Au_{11}, Fe_4/Au_{10}, and Fe_6/Au_8.

A phenomenological description of the magnetic coupling proposed to explain the experimental observations gives the expression for the interlayer coupling energy per unit area A as[1]

$$\frac{E}{A} = -J_1 \cos\Theta - J_2 \cos^2\Theta, \tag{1}$$

where Θ is the angle between the magnetization directions of two adjacent Fe layers, and J_1 and J_2 are the bilinear and biquadratic coupling constants, which describe the type and the strength of the coupling. If the term with J_1 dominates, then the coupling is ferromagnetic or antiferromagnetic. Positive values of the coupling constant J_1 favor parallel alignment of the magnetizations and negative values favor the antiparallel alignment. If the term with J_2 dominates and is negative, we can observe the 90°-configuration.

The bilinear coupling constant can be estimated from the total-energy calculations as the difference in energy between the antiferromagnetic and ferromagnetic arrangement of the Fe layers:

$$J_1 = \frac{1}{2A}(E_{AF} - E_F). \tag{2}$$

In Fig. 1 the calculated bilinear coupling constant J_1 is shown and compared with two measured values for Fe_2/Au_2 and Fe_4/Au_4. The preferred alignment of the magnetization,

and hence the sign of J_1, oscillates as the thickness of the spacer layer varies. With increasing spacer thickness, the positive J_1 constant changes sign for 6 or 8 Au monolayers. This effect depends on the Fe layer thickness.

The only ferromagnetic alignment was already observed experimentally for low periodicity Fe_n/Au_n multilayers.[11] The strength of the measured interlayer coupling is in accordance with our findings for Fe_2/Au_2 and Fe_4/Au_4. The experimental studies of the Fe/Au/Fe trilayers on a GaAs substrate showed the ferromagnetic ordering in structures with Au spacer thicknesses up to eight Au monolayers.[1]

The other total-energy calculations, based on the tight-binding model and the screened Korringa-Kohn-Rostoker (KKR) method, were performed for systems with much thicker layers of either Au spacer[5] or Fe.[6] Although it is not possible to make a quantitative comparison between our findings and the aforementioned results, the changes of the magnetic ordering with the spacer thickness seem to be well-reproduced.

In some systems, the peculiar effect of the orthogonal alignment of magnetic moments, rather than collinear alignment, is observed.[20,21] In these cases, the higher order coupling terms, Eq. (2), have to be considered. However, higher order terms are usually much smaller than the bilinear term, so in this report they were neglected. Generally, the biquadratic coupling constant could be calculated from the total-energy calculations as is shown for the Fe/FeSi system in Ref. 22.

B. Interface magnetism

Previous *ab initio* calculations indicate that the ferromagnetic ordering of one Fe layer is preferred in all Fe/Au structures, independently of the Fe layer thickness.[23] Thus we limited our calculations to the configurations with parallel and antiparallel ordering of adjacent Fe layers which are ferromagnetically ordered. The profiles of magnetic moments calculated for Fe_2/Au_{10} and Fe_4/Au_8 are shown in Fig. 2. Since the Au monolayers are only slightly polarized by the neighboring Fe layers, the magnetic moments of Au atoms are multiplied by a factor of 40. The calculations show that the magnitudes of Fe moments in ferro- and antiferromagnetic states do not differ.

The calculated magnetic spin moments for Fe_n/Au_m systems with different Fe and Au layer thicknesses are summarized in Table II. We observe that the calculated magnetic moments of Fe atoms are larger than the bulk value $2.08\mu_B$ calculated previously using the same method.[19] For Fe atoms placed at the interfacial sites, the values of $2.66\mu_B$, $2.74\mu_B$, and $2.78\mu_B$ were found in multilayers with two, four, and six Fe monolayers, respectively. For the interior Fe monolayers of Fe_4/Au_m or Fe_6/Au_m, the magnetic moments of Fe atoms are reduced to about $2.5\mu_B$. The magnetic moments of interfacial Au are much lower. Their values increase dramatically with the Fe layer thickness, from 0.038 for Fe_2/Au_m up to $0.062\mu_B$ for Fe_4/Au_m. Then, they stay almost unchanged for Fe_6/Au_m. The value of the magnetic moment of subsequent Au monolayers drops by an order of magnitude, and consequently it is zero at the middle Au monolayer for multilayers larger than three Au monolayers.

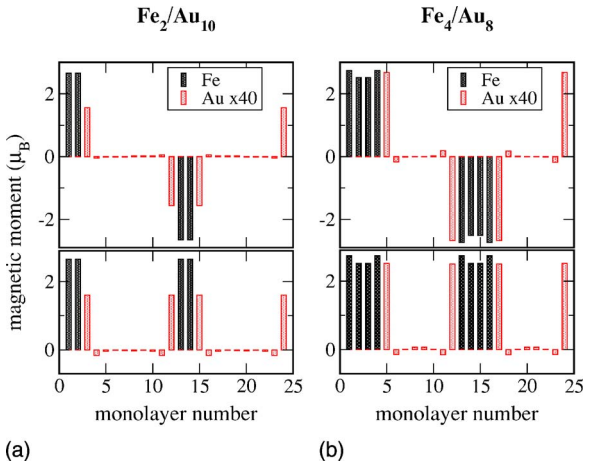

FIG. 2. (Color online) Distribution of magnetic moments in the magnetic unit cell of Fe_2/Au_{10} and Fe_4/Au_8, for two magnetic configurations with (a) antiferromagnetic order and (b) ferromagnetic order of Fe layers separated by Au monolayers. The induced Au moments have been enlarged by a factor of 40.

The presented results are in accordance with previous findings,[19,24,25] however, our calculations are extended to multilayers with a thicker Au spacer.

IV. SUMMARY

We have estimated the exchange coupling between magnetic Fe layers when separated by nonmagnetic Au spacer layers using the *ab initio* approach, with the electronic structure calculated self-consistently. Previous asymptotic calculations[13] and *ab initio* calculations[4–6] have ignored the electron-electron interaction in the spacer. During the structure optimization, we do not fix either the lattice constants nor the distances between layers, allowing the preferred

TABLE II. The calculated magnetic spin moments (μ_B) of the interfacial Au and Fe atoms placed in different atomic layers of Fe_n/Au_m systems.

Structure	Interfacial Au monolayer	Interfacial Fe monolayer	Interior Fe monolayers	
Fe_2/Au_2	0.038	2.664		
Fe_2/Au_4	0.038	2.663		
Fe_2/Au_6	0.037	2.666		
Fe_2/Au_8	0.039	2.662		
Fe_2/Au_{10}	0.039	2.659		
Fe_4/Au_2	0.063	2.739	2.485	
Fe_4/Au_4	0.062	2.741	2.487	
Fe_4/Au_6	0.061	2.746	2.491	
Fe_4/Au_8	0.061	2.742	2.488	
Fe_6/Au_2	0.063	2.782	2.465	2.501
Fe_6/Au_4	0.061	2.782	2.472	2.549
Fe_6/Au_6	0.065	2.777	2.408	2.444
Fe_6/Au_8	0.066	2.782	2.431	2.478

atomic configuration for ferro- and antiferromagnetic arrangement of two adjacent Fe layers to be found. The difference between the calculated total energies for parallel and antiparallel ordering of Fe_n/Au_m antiferromagnetic unit cells ranges from 1 to 20 meV per supercell. It is a very subtle difference in comparison to the total energy of a supercell which is on the order of hundreds of eV.

The strength of the bilinear coupling constant for Fe_2/Au_2 and Fe_4/Au_4 determined from our calculations is in very good agreement with known experimental data.[11] The strong ferromagnetic coupling observed for Fe_n/Au_2 is probably due to the additional contribution to the magnetic coupling generated by the direct interaction of the interfacial Fe atoms, which are separated only by two monolayers of Au. For increasing spacer thicknesses, the ferromagnetic coupling quickly decreases.

The employed method gives the opportunity of determining the magnetic moment distribution over the multilayer. The results show that the magnetic moments of the Fe and Au interface do not depend on the spacer thickness. The magnitude of magnetic moments induced on the interfacial Au layer is an order of magnitude smaller than that on Fe atoms. Moreover, for Fe_2/Au_m structures the Au interfacial magnetic moment is almost two times smaller than that for Fe_4/Au_m and Fe_6/Au_m.

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[1] P. Grünberg, J. Phys.: Condens. Matter **13**, 7691 (2001).
[2] M. D. Stiles, J. Magn. Magn. Mater. **200**, 322 (1999).
[3] J. Unguris, R. J. Celotta, and D. T. Pierce, Phys. Rev. Lett. **79**, 2734 (1997).
[4] L. Szunyogh, B. Újfalussy, P. Weinberger, and C. Sommers, Phys. Rev. B **54**, 6430 (1996).
[5] A. T. Costa, Jr., J. d'Albuquerque e Castro, and R. B. Muniz, Phys. Rev. B **56**, 13697 (1997).
[6] J. Opitz, P. Zahn, J. Binder, and I. Mertig, Phys. Rev. B **63**, 094418 (2001).
[7] D. T. Dekadjevi, P. A. Ryan, B. J. Hickey, B. D. Fulthorpe, and B. K. Tanner, Phys. Rev. Lett. **86**, 5787 (2001).
[8] T. L. Monchesky, A. Enders, R. Urban, K. Myrtle, B. Heinrich, X.-G. Zhang, W. H. Butler, and J. Kirschner, Phys. Rev. B **71**, 214440 (2005).
[9] K. Takanashi, S. Mitani, H. Fujimori, K. Sato, and Y. Suzuki, J. Magn. Magn. Mater. **177-181**, 1199 (1998).
[10] L. Uba, S. Uba, V. N. Antonov, A. N. Yaresko, T. Ślęzak, and J. Korecki, Phys. Rev. B **62**, 13731 (2000).
[11] A. Yoshihara, J. T. Wang, K. Takanashi, K. Himi, Y. Kawazoe, H. Fujimori, and P. Grünberg, Phys. Rev. B **63**, 100405(R) (2001).
[12] J. Unguris, R. J. Celotta, and D. T. Pierce, J. Appl. Phys. **75**, 6437 (1994).
[13] P. Bruno and C. Chappert, Phys. Rev. Lett. **67**, 1602 (1991).
[14] P. Bruno, Phys. Rev. B **52**, 411 (1995).
[15] M. D. Stiles, J. Appl. Phys. **79**, 5805 (1996).
[16] G. Kresse and J. Hafner, Phys. Rev. B **47**, 558 (1993); **49**, 14251 (1994).
[17] G. Kresse and J. Furthmuller, Software VASP (Vienna, 1999); Phys. Rev. B **54**, 11169 (1996); Comput. Mater. Sci. **6**, 15 (1996).
[18] P. E. Blöchl, Phys. Rev. B **50**, 17953 (1994); G. Kresse and D. Joubert, *ibid.* **59**, 1758 (1999).
[19] M. Sternik, K. Parlinski, and J. Korecki, Phys. Rev. B **74**, 195405 (2006).
[20] J. C. Slonczewski, Phys. Rev. Lett. **67**, 3172 (1991); J. Appl. Phys. **73**, 5957 (1993).
[21] E. E. Fullerton and S. D. Bader, Phys. Rev. B **53**, 5112 (1996).
[22] P. T. Jochym, K. Parlinski, and A. M. Oleś, Phys. Rev. B **73**, 224411 (2006).
[23] J.-T. Wang, L. Zhou, D.-S. Wang, and Y. Kawazoe, Phys. Rev. B **62**, 3354 (2000).
[24] M. Böhm and U. Krey, J. Magn. Magn. Mater. **192**, 27 (1999).
[25] J.-T. Wang, Z.-Q. Li, Q. Sun, and Y. Kawazoe, J. Magn. Magn. Mater. **183**, 42 (1998).