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First-principles Calculations of Magnetic Anisotropy of Fe and Co Films, Separated by Nonmagnetic Metallic Interlayers

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Abstract—The magnetic characteristics of Co and Fe monolayer films on Cu and Pt surfaces have been numerically calculated using the VASP software package. Dependences of a difference in the total energies of the antiferromagnetic and ferromagnetic configurations on the convergence parameters and nonmagnetic-material thickness are calculated. The values of magnetic anisotropy and atomic magnetic moments in the Co/Cu/Co, Fe/Pt/Fe, Co/Pt/Co, and Pt/Co/Cu/Co/Pt structures are determined in dependence of the surface-face orientation. The reorientation phenomenon in the Co/Cu/Co structure (when anisotropy of Co films, parallel to the surface plane, is changed to perpendicular anisotropy because of introduction of an ultrathin Pt film into the structure) is confirmed for the (110) and (111) faces.

Keywords: magnetic anisotropy, ultrathin magnetic films

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1. INTRODUCTION

Lately, the properties of thin magnetic films attract large interest of researchers. Thin magnetic films are used as the main material for magnetic recording devices. In view of rapid development of magnetic recording, calculation of the magnetic characteristics of Co and Fe films became an urgent problem of solid-state physics.

One of the actual problems is the study of magnetic properties of multilayer structures [1], exhibiting giant magnetoresistance (GMR), because they are widely applied for read/write heads of hard drives and storage and spintronic devices, the properties of which depend strongly on magnetic anisotropy (MA). Multilayer GMR structures consist of ferromagnetic (FM) layers, separated by nonmagnetic layers. Due to the oscillating RKKY interaction, the interlayer-exchange constant is varied at a change in the distance between FM layers. By varying the thickness of nonmagnetic material between two magnetic layers, one can change interaction from FM to antiferromagnetic (AFM). Therefore, it is important to predict theoretically at which nonmagnetic-layer thickness the AFM configuration is more energetically favorable.

Since the current magnetic-storage density approaches the superparamagnetic limit, it is believed that higher surface densities can be reached using perpendicular recording because of lower superparamagnetic limit in the magnetic media with perpendicular anisotropy [2]. While most systems exhibit MA in the film plane, recently, the multilayer structures, consisting of Co/Pt layers with perpendicular MA, attracted much attention due to their potential for ultrahigh-density data recording [3–6]. Perpendicular anisotropy in these systems is proportional to the interface area [7]. If the Co layer thickness is smaller than the critical value (1 nm), surface anisotropy exceeds shape anisotropy and a multilayer system becomes perpendicularly magnetized [8].

To calculate the MA energy, one should estimate a difference between the total energies of the magnetic material for different magnetization orientations. It is a very complex numerical problem because of stronger dependence on the calculation parameters. Magnetic moments and MA for the Co/Pt(111) and Fe/Pt(111) systems were studied in, e.g., [9, 10]. All these calculations are based on the Green's function approach within the completely relativistic screened Kohn–Korringa–Rostoker method and approximations of local density and idealized geometry, which reproduces the bulk structure of the substrate.

We analyzed MA of monolayer Fe and Co films on Cu and Pt within the SDFT formalism and calculated the self-consistent band structure using the VASP program package. Differences in the results of calculating MA using different methods were analyzed in [11]. We investigated not only FM states (as in [9, 10]) but also the AFM configurations with complete relaxation of the entire system, consisting of an adatom and a substrate. In addition, in contrast to the most popular calculations for the (111) surface face, we investigated other orientations of the surface faces (in particular

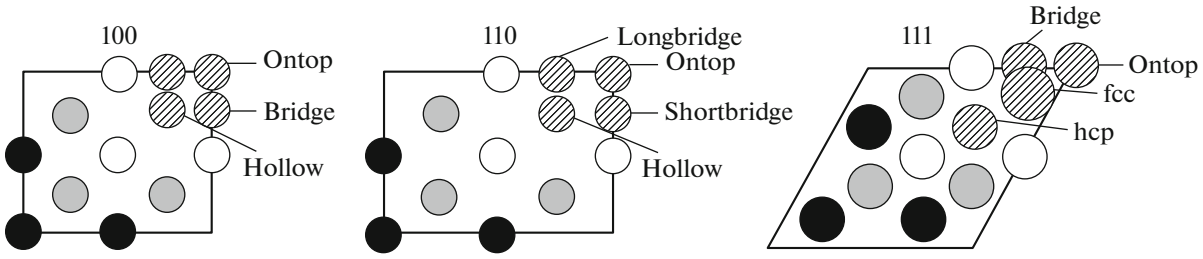

Fig. 1. Configurations of adatom positions depending on the surface-face orientation.

(100), because it yields a square surface cell, which is used in our Monte Carlo calculations).

The results of experiment [12] and our Monte Carlo calculations [13, 14] yielded that the case of perpendicular anisotropy induces a useful increase in magnetoresistance in a structure with FM films with thicknesses of $N < 11$ monolayers. Therefore, the purpose of this study was to investigate using ab initio calculations the experimentally observed phenomenon of reorientation of Co films in the Co/Cu/Co structure, when anisotropy changes orientation from parallel with respect to the film plane to perpendicular because of sputtering of an ultrathin Pt film onto the structure.

2. MODEL AND TECHNIQUES

The first-principles calculations of the electronic and crystal structures of magnetic materials are based on the spin-density-functional theory. The multilayer structures were simulated using the VASP software package [15] according to the PAW method with the PBE version of the generalized gradient approximation (GGA). Magnetization was taken into account in collinear and noncollinear versions, in which the magnetic moment is set by a scalar value and a vector, respectively. Integration in the first Brillouin zone was performed using a k-mesh, constructed according to the Monkhorst–Pack method. The plane-wave cutoff energy was chosen to be $E_{cut} = 500$ eV at the vacuum-layer thickness of 5 Å. The k-mesh sizes were varied from $5 \times 5 \times 1$ to $55 \times 55 \times 1$.

Table 1. Total energies E (in eV) for the Fe/Pt/Fe and Co/Pt/Co structures

Face	Adatom position	Fe/Pt	Co/Pt
100	Ontop	–42.7	–39.60
	Hollow	**–45.1**	**–42.34**
	Bridge	–44.0	–41.23
110	Ontop	–41.3	–37.20
	Hollow	**–45.3**	**–42.71**
	Longbridge	–43.6	–40.56
	Shortbridge	–42.1	–39.24
111	Ontop	–44.2	–41.21
	hcp	**–45.3**	–42.00
	fcc	–44.8	**–42.61**
	Bridge	–45.3	–42.60

A film atom can be adsorbed onto the substrate in different configurations, shown in Fig. 1. Therefore, the values of the total supercell energy were calculated in dependence on the surface face and adatom position for Fe and Co monolayer films on the Pt substrate with a thickness of 5 monolayers (the obtained results are presented in Table 1).

One can conclude that most energetically favorable position of both Fe and Co atoms on the Pt substrate is interstitial arrangement of adatoms (hollow site for the 100 and 110 faces and hcp or fcc sites for the 111 face). In further studies, this system was a nonmagnetic metallic plate with the surface-face orientation with low values of the Miller index. For conservation of inversion symmetry, FM films were adsorbed onto both sides of this plate in the interstitial sites.

The multilayer structure was simulated using a periodic supercell with the lattice constant, matched to the substrate and calculated taking into account optimization of the lattice parameters. The calculated equilibrium lattice parameters ($a = 3.6367(5)$ Å for Cu and $a = 3.87125(3)$ Å for Pt) are close to the experimental values ($a = 3.6153$ Å for Cu [16] and $a = 3.9158$ Å for Pt [17]).

3. ANALYSIS OF EXPEDIENCY OF DIFFERENT SPIN CONFIGURATIONS

First, calculations were carried out for a monolayer FM film on the Pt(100) surface in dependence of the convergence parameters and the nonmagnetic-interlayer thickness. Complete relaxation of all supercell layers was performed.

The total energies for the FM and AFM spin configurations were calculated (Fig. 2). We introduced the value $\Delta E = E_{AFM} - E_{FM}$, equal to a difference in the total energies of the systems, when the atomic magnetic moments in the films at the plate sides are

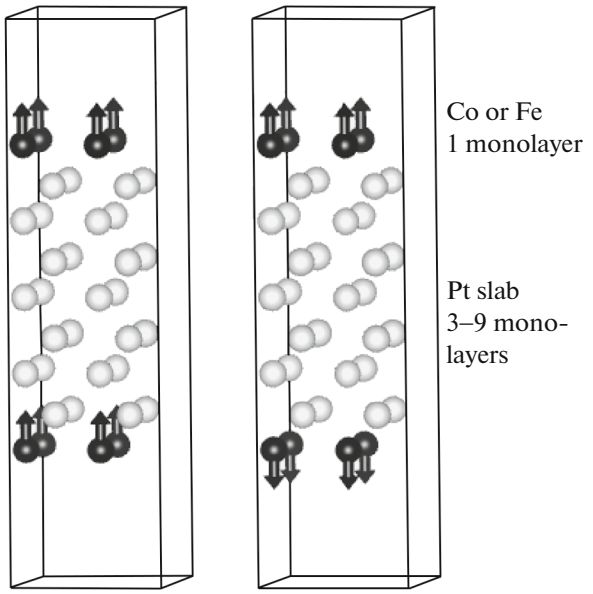

Fig. 2. Crystal structure of a supercell and orientation of atomic magnetic moments in the film.

directed in the opposite (AFM configuration) and identical (FM configuration) directions. At $\Delta E < 0$ and $\Delta E > 0$, more energetically favorable is the AFM and FM orientation, respectively.

Study of convergence over the number of k-points (mesh-partition points at integration in the first Brillouin zone) yielded that convergence for ΔE (Fig. 4) is much worse than for the total energy (Fig. 3). Therefore, one should use more k-points (mesh size of about $50 \times 50 \times 1$), whereas a sufficient mesh size for calculation of the adsorption energy and magnetic moments is about $15 \times 15 \times 1$. Insufficient number of k-points may even lead to a change in the ΔE sign (Fig. 6a).

However, one should note that a significant increase in the number of k-points increases sharply the computing resources: calculation time and, primarily, memory (Fig. 5).

Figure 6 shows the results of calculating the dependence of the ΔE values for the Fe and Co films, which are adsorbed at two sides of the Pt(100) plate, on the plate thickness.

Calculations show that most energetically favorable orientation for Co films with thicknesses of 1 and 2 monolayers for all considered thicknesses of the Pt plate is the FM orientation, which is changed to AFM only for a three-layer Co film at the Pt thickness of more than 5 monolayers.

AFM orientation is favorable for a monolayer Fe film for all considered Pt thicknesses. However, the ΔE value for Fe is much smaller than for Co (~1 meV).

Thus, one can conclude that occurrence of the AFM interaction between two magnetic layers

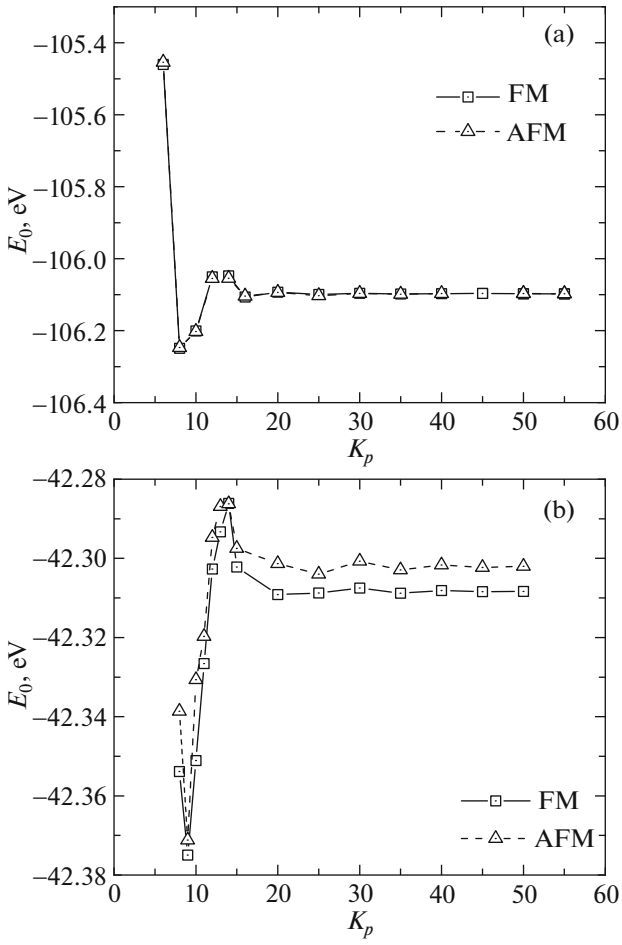

Fig. 3. Convergence of the total energy in dependence of the number of k-points for (a) Fe/Pt/Fe and (b) Co/Pt/Co.

depends on the thickness of not only the nonmagnetic material but also magnetic layers.

4. CALCULATIONS OF THE MAGNETIC ANISOTROPY

Let us consider multilayer structures with layer thickness of 3 mL (Fig. 7) in dependence of the magnetization direction and the surface-face orientation.

The MA energy $E_{MA} = E_{\perp} - E_{\parallel}$ is a difference between the total energies of the systems, the magnetization in which is directed perpendicular and parallel to the surface plane, respectively. At $E_{MA} < 0$, the orientation of magnetic moments perpendicular to the surface, which corresponds to easy axis, is more energetically favorable. At $E_{MA} > 0$, the orientation of magnetic moments parallel to the surface, which corresponds to easy plane, is more energetically favorable.

The results of calculating a difference in the total energies for different spin configurations of the structures based on Co films with nonmagnetic Pt and

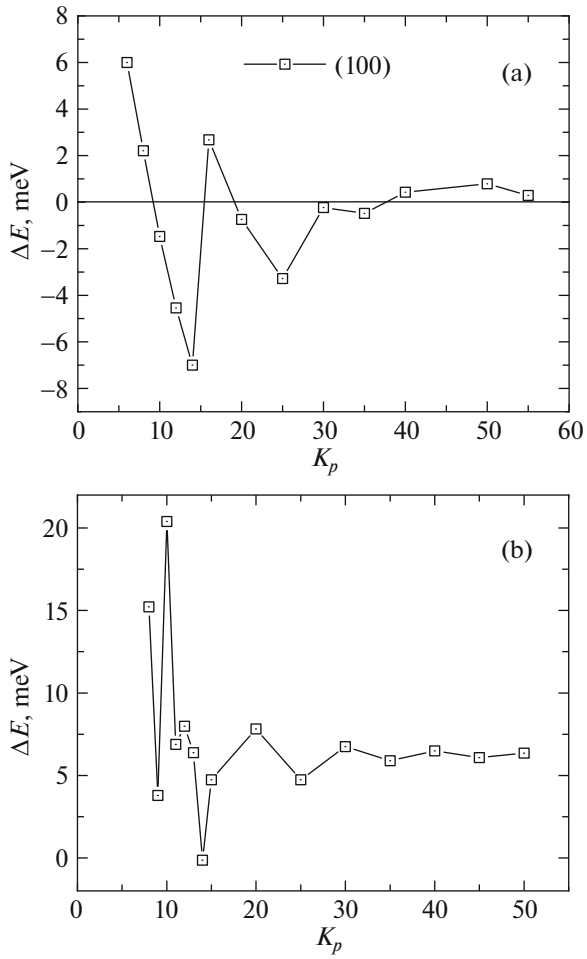

Fig. 4. Convergence of a difference of the energies of AFM and FM configurations in dependence of the number of k-points for (a) Fe/Pt/Fe and (b) Co/Pt/Co.

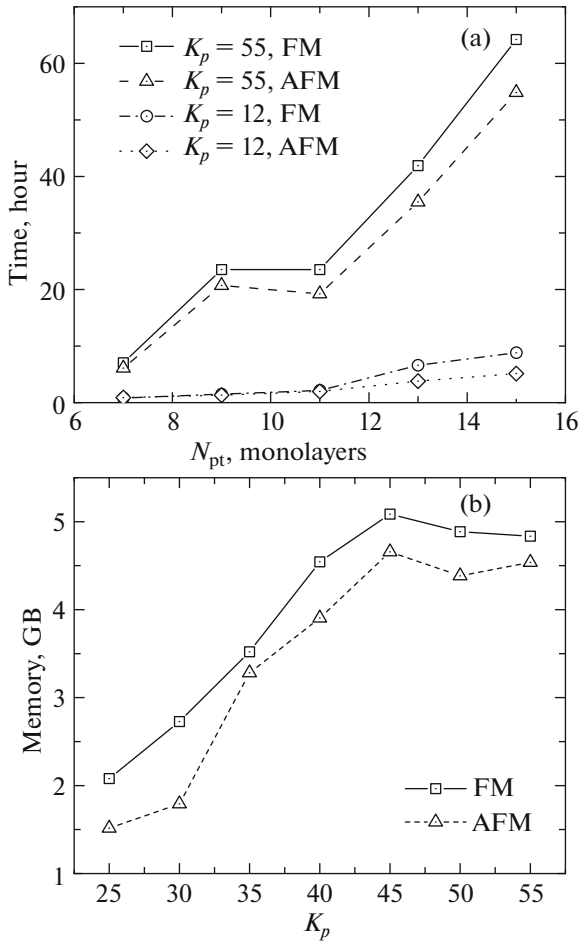

Fig. 5. Dependences of (a) calculation time on the Pt plate thickness at the mesh sizes of 12 ×12 × 1 and 55 × 55 × 1 and (b) required core memory on the number of k-points of the AFM and FM configurations for Fe/Pt/Fe.

Cu interlayers and a Pt film, deposited over the Co/Cu/Co structure, are listed in Tables 2, 3.

The results of calculating a difference in the energies of the AFM and FM configurations ΔE for Co films with Cu interlayers showed that the FM and AFM configurations are more favorable for the (100) face and for the rest faces, respectively. Deposition of an additional upper Pt layer changes the ΔE sign only for the (111) face. For Co films with Pt interlayers, the FM configuration is more favorable for almost all considered cases.

Table 2. Calculated differences in the energies of the AFM and FM configurations ΔE in dependence of the surface-face orientation and magnetization direction

Face	Alignment type	ΔE, meV		
		Co/Pt/Co	Co/Cu/Co	Pt/Co/Cu/Co/Pt
100	∥	12.344	9.466	16.000
	⊥	12.889	6.631	17.034
110	∥	5.592	−31.784	−30.755
	⊥	8.426	−31.585	−28.443
111	∥	4.965	−7.160	3.448
	⊥	−1.406	−6.854	3.077

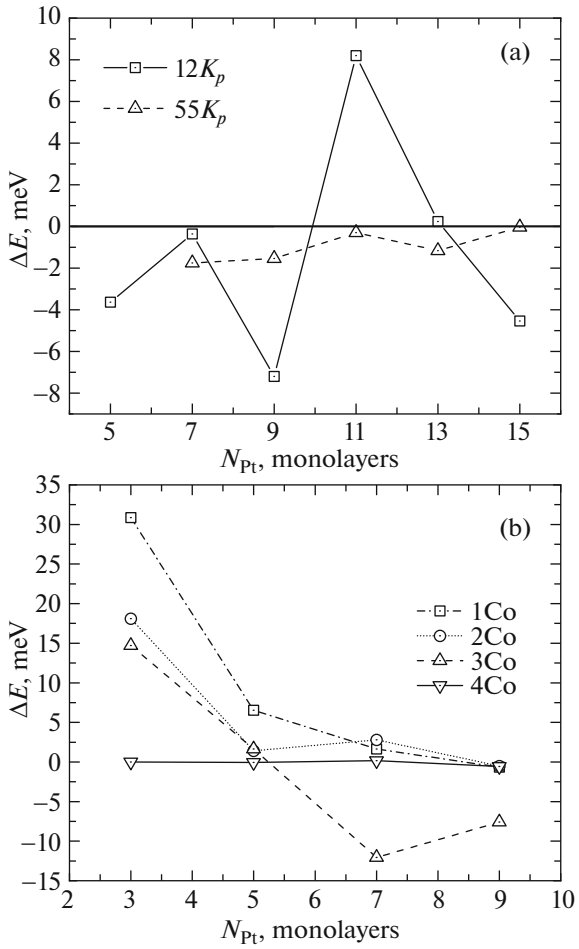

Fig. 6. Dependence of a difference in the energies of the AFM and FM configurations on the Pt plate thickness for (a) Fe/Pt/Fe and (b) Co/Pt/Co at different mesh sizes.

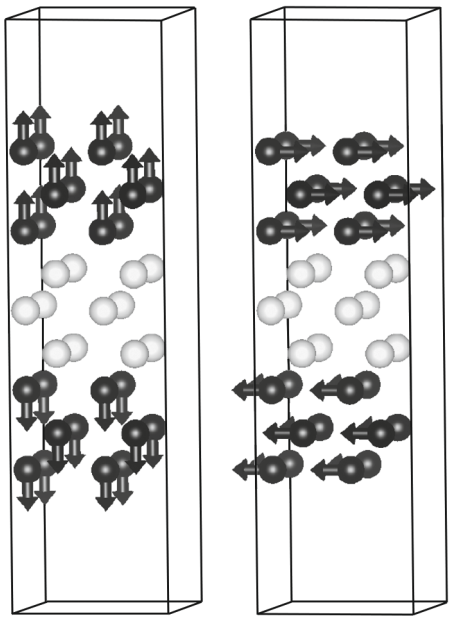

Fig. 7. Crystal structure of a supercell and orientation of the atomic magnetic moments in the film.

It was found experimentally [12] that the Co/Cu structure is characterized by weak easy-plane MA with magnetization, oriented in the film plane. However, deposition of ultrathin Pt films onto the Co film changes the anisotropy of the Pt/Co/Cu/Co/Pt structure to the easy-axis type with magnetization, oriented perpendicular to the film plane. This fact is also confirmed by our calculations. For the Co/Cu/Co structure, $E_{MA} = E_\perp - E_\parallel > 0$, which corresponds to the easy-plane MA (except very small values $E_{MA} = -0.05$ eV at energetically unfavorable FM configurations of the (110) and (111) faces). As was reported in [12], additional measurements, performed by Kohlhepp et al. (1992) and den Broeder et al. (1991), confirmed that the Co/Cu(111) system exhibits weak anisotropy, oriented perpendicular to the interface of ~0.1 mJ/m^2.

It was confirmed experimentally [12] that the easy-axis anisotropy with magnetization, oriented perpendicular to the film plane, arises in the Pt/Co/Cu/Co/Pt and Co/Pt/Co structures for the FM configuration of the (110) face and the AFM configuration of the (111) face, respectively, because perpendicular MA in these structures is observed specifically for the (111) face.

Table 3. Calculated MA energies $E_{MA} = E_\perp - E_\parallel$ in dependence of the surface-face orientation

Face	Spin configuration	E_{MA}, meV		
		Co/Pt/Co	Co/Cu/Co	Pt/Co/Cu/Co/Pt
100	FM	0.965	3.204	1.961
	AFM	1.510	0.369	2.995
110	FM	0.993	–0.050	–2.205
	AFM	3.827	0.149	0.107
111	FM	0.463	–0.044	–0.034
	AFM	–5.908	0.262	–0.405

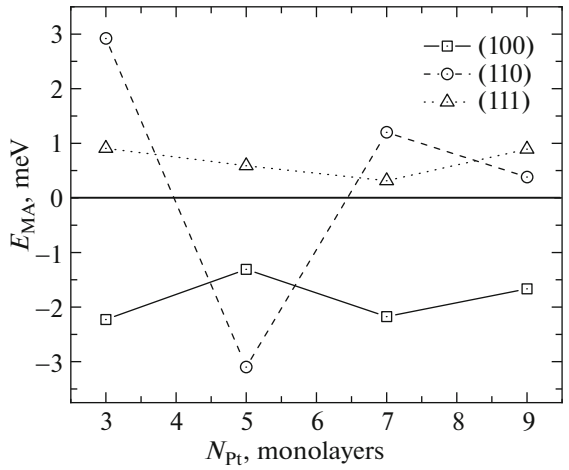

Fig. 8. Dependence of MA for the 1Fe/Pt/1Fe system on the Pt-interlayer thickness for different surface-face orientations.

The results of calculating the MA energies yielded that, at the (100) surface-face orientation, all considered structures based on Co films with nonmagnetic Pt and Cu interlayers and a Pt film, deposited over the Co/Cu/Co structure, are characterized by the easy-plane MA with magnetization, oriented in the film plane.

The results of calculating the magnetic moments of Co atoms for the systems under consideration are shown in Table 4. Co atoms in the Co/Pt/Co and Co/Cu/Co structures have the largest and the least magnetic moments, respectively. The mean magnetic moment of Co atoms (Table 4) is larger for the structure with a Pt interlayer and barely depends on the spin configurations (it changes by ~$0.01\mu_B$).

Let us now investigate the structures based on Fe films in dependence of the surface-face orientation. The results of calculating the MA energies and magnetic moments for monolayer Fe films with Pt interlayers are presented in Fig. 8 and Table 5. One can see that perpendicular anisotropy for monolayer Fe films with Pt interlayers arises only for the (100) face for all Pt-plate thicknesses and for the (110) face for the Pt-plate thickness of 5 monolayers. For the (111) face, easy-plane anisotropy with $E_{MA} = E_\perp - E_\parallel \approx 0.5$–1.0 meV is more energetically favorable for all nonmagnetic-interlayer thicknesses.

Our calculations for the (111) face were confirmed by other authors. Adsorption of Fe clusters of different geometry (up to a monolayer on the Pt(111) surface face) was investigated in [19]. The calculations were performed within the Kohn–Korringa–Rostoker (KKR) formalism [18] at the Pt substrate thickness of 37 monolayers with the system relaxation disregarded (k-mesh size of 100 × 100 × 1). For all clusters of the Fe/Pt(111) system under study, easy-axis MA dominates; however, easy-plane anisotropy with $E_{MA} = E_\perp - E_\parallel = 0.26$ meV dominates for a monolayer. The calculations of a single Fe monolayer on Pt(111), which were carried out in [10], yielded in-plane MA with $E_{MA} = 0.71$ meV, whereas a single Fe adatom on Pt(111) is oriented strictly perpendicular.

The results of calculating the MA energy and magnetic moments for Fe films, separated by Pt interlayers with thicknesses of all materials of 3 monoatomic layers, are presented in Table 6.

The calculations showed that perpendicular MA is observed for all surface-face orientations of Fe films

Table 4. Calculated mean magnetic moments of Co atoms μ, μ_B and magnetic moments of Co atom μ_1, μ_B, which is the closest to the nonmagnetic middle interlayer

Face	Configuration type		Co/Pt/Co		Co/Cu/Co		Pt/Co/Cu/Co/Pt	
			μ, μ_B	μ_1, μ_B	μ, μ_B	μ_1, μ_B	μ, μ_B	μ_1, μ_B
100	FM	∥	1.765	1.851	1.724	1.885	1.751	1.836
		⊥	1.764	1.850	1.721	1.871	1.753	1.835
	AFM	∥	1.766	1.847	1.721	1.870	1.752	1.834
		⊥	1.765	1.847	1.722	1.872	1.751	1.831
110	FM	∥	1.854	1.889	1.732	1.871	1.833	1.884
		⊥	1.850	1.893	1.732	1.867	1.834	1.892
	AFM	∥	1.848	1.896	1.721	1.854	1.835	1.893
		⊥	1.847	1.894	1.721	1.855	1.835	1.893
111	FM	∥	1.814	1.853	1.701	1.773	1.805	1.858
		⊥	1.814	1.854	1.701	1.775	1.805	1.856
	AFM	∥	1.812	1.850	1.702	1.780	1.805	1.856
		⊥	1.812	1.852	1.701	1.774	1.806	1.857

Table 5. Calculated μ, μ_B values of a monolayer Fe film at the Pt plate thickness of 9 monolayers in comparison with the calculation results obtained using the KKR formalism at the Pt substrate thickness of 37 monolayers [19] and on a semi-infinite Pt surface [10]

Type	(111)	(110)	(100)
∥	2.98 3.018 [10]	3.15	3.06
⊥	2.97 2.92 [19] 3.016 [10]	3.15	3.05

Table 6. Calculated MA energies E_{MA} (in meV) and magnetic moments μ, μ_B of Fe atoms and surface Pt layer for different surface-face orientations for the 3Fe/3Pt/3Fe system

Face		E_{MA}, meV	μ_{Pt}	μ_{Fe_1}	μ_{Fe_2}	μ_{Fe_3}
100	⊥	−1.57	0.293	2.752	2.399	2.906
	∥		0.283	2.748	2.402	2.893
110	⊥	−1.10	0.358	2.842	2.800	2.848
	∥		0.357	2.842	2.801	2.848
111	⊥	−1.45	0.273	2.833	2.588	2.852
	∥		0.266	2.835	2.592	2.859

with a thickness of 3 monoatomic layers and only for the (100) face orientation in the case of a monoatomic film.

CONCLUSIONS

To conclude, we should note that ab initio calculations of MA of Co and Fe films separated by Cu and Pt plates were performed in dependence of the convergence parameters and plate thickness. Convergence analysis over the number of k-points yielded that convergence for ΔE is much worse than for the total energy. Thus, one should use more k-points (mesh size of about $50 \times 50 \times 1$).

For Co films with thicknesses of 1 and 2 monolayers, FM orientation is more energetically favorable for all considered thicknesses of the Pt plate and is changed to AFM only for a three-layer film with Pt thickness of more than 7 monolayers. For a monolayer Fe film, AFM orientation is favorable for all considered Pt thicknesses.

The MA energies and magnetic moments for atoms in the Fe/Pt/Fe, Co/Cu/Co, Co/Pt/Co, and Pt/Co/Cu/Co/Pt structures with thicknesses of each metal of 3 monolayers were determined in dependence of the surface-face orientation. The Co/Cu/Co structure is characterized by easy-plane MA, except very weak perpendicular anisotropy at energetically unfavorable FM configurations of the (110) and (111) faces.

However, if one deposits ultrathin Pt films onto the Co films for the (111) face and FM configuration of the (110) face, the obtained Pt/Co/Cu/Co/Pt structure is characterized by easy-axis anisotropy. For the Co/Pt/Co structure, perpendicular anisotropy is more energetically favorable for the AFM configuration of the (111) face.

For the (100) orientation of the surface face, all considered structures based on Co films with nonmagnetic Pt and Cu interlayers and Pt film deposited over the Co/Cu/Co structure are characterized by the easy-plane MA.

Perpendicular anisotropy was observed at all surface-face orientations for Fe films with thickness of 3 monoatomic layers and only at the (100) face for a monoatomic film.

Our conclusions are in agreement with the experimental results, obtained in [12] and [10, 19]. Our results can be applied at numerical Monte Carlo simulation of nonequilibrium behavior of multilayer magnetic superstructures [20].

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CONFLICT OF INTEREST

The authors declare that they have no conflicts of interest.

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