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Ab Initio Calculations of Magnetic Anisotropy in Fe/Pt/Fe Heterostructures Taking into Account Surface Relaxation

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Abstract—Numerical studies on the effect of surface relaxation, namely, the equilibrium change in the interplanar distance, exerted on the values of the magnetic and energy characteristics of a Fe/Pt/Fe system have been performed. The contribution of spin-orbit coupling to the value of magnetic anisotropy energy is studied as a function of ferromagnetic film thickness.

Keywords: surface relaxation, magnetocrystalline anisotropy, spin-orbit coupling, ab initio calculation

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INTRODUCTION

The spin density functional theory (SDFT) is a rather accurate approach to the static electron many-body problem for an equilibrium electron gas, which is widely used in a vast number of studies to describe different properties of the ground state, both for magnetic and nonmagnetic solids.

One of the properties considered in this paper is magnetocrystalline anisotropy, i.e., the tendency of the system towards the orientation of its magnetization predominantly in one direction with respect to the crystal lattice.

Such a property is one of the most important of magnetic materials, since it also attractive for technological applications in instrumentation and information technologies, as well as spintronics. However, magnetocrystalline anisotropy is also of interest for fundamental physics.

Ab initio calculations of the magnetic anisotropy energy (E_{MA}) are particularly difficult. First, there are substantial technical difficulties, since obtaining E_{MA} means that the difference between the total energy for different magnetization orientations should be calculated with great accuracy. Second, factors difficult to control, including many-particle effects beyond the local density approximation (LDA) [1–3], could affect the calculated E_{MA} value.

Sequentially alternating conductive layers made of ferromagnetic and nonmagnetic materials based on a Fe/Pt/Fe multilayer structure make it possible to achieve giant magnetoresistance effects. The magnetic properties of ferromagnetic Fe films and the behavior of exchange-coupled structures based on these films are being intensively studied.

Relaxation studies show that the presence of an inhomogeneous pattern of interatomic distances causes a significant change in the E_{MA} value and can also be the reason for the reorientation of magnetization in atoms. Papers by different authors confirm this. In [3], the relaxation effect on E_{MA} for clusters with a face-centered cubic (fcc) transition metal lattice has been studied. In [4], the relaxation effect on E_{MA} has been studied for the case of Co wires at the edge of a Pt(111) step. In both papers, an improvement in the quantitative description of E_{MA} is observed when surface relaxation is taken into account.

Studies of Fe films on the surfaces of different nonmagnetic metals, including Pt, is of great interest to experimenters, because in order to investigate the main structural properties and their relationship with the magnetic properties, it is necessary to use an epitaxial film, since the crystallographic orientation can be controlled by the orientation of the substrate. For example, Fe/Pt epitaxial films have been prepared on the surface of single-crystal substrates [4–8]. Studies have shown that both the film thickness and choice of substrate material, as well as the orientation of the surface face, can affect the predominant type of E_{MA}.

The authors of [9] have studied a Fe film on an Au surface depending on the film thickness, for the case of (100) face using the Korringa–Kohn–Rostoker (KKR) formalism. For example, for the case of monolayer film thicknesses and two atomic layers, perpendicular anisotropy predominates, whereas with a

greater thickness the anisotropy exhibits a planar character. In addition, the type of predominant anisotropy can be affected by the substrate onto the surface of which the film is deposited.

Studies performed by the authors of [10] have shown that the in-plane anisotropy always prevails in the case of Fe films on the surface of W(110), whereas on the surface of Cr(110), for the case of small film thicknesses, an easy axis type of anisotropy is observed, and when the thickness exceeds three monolayers, prevalent in-plane anisotropy is revealed. Calculations based on the Green's function approach, a completely relativistic KKR method involving the local density approximation and an idealized geometry reproducing the bulk structure of the substrate, have shown the predominance of in-plane anisotropy for the case of Fe monolayer deposited onto the Pt(111) surface [11].

The authors of [1] studied the effect of surface relaxation on the magnetic anisotropy energy for the case of the (100) face using a theoretical ab initio approach. The results have shown that surface relaxation can affect not only the E_{MA} value, but also the predominant type of anisotropy. Thus, for the Fe/Pt system without relaxation, in-plane anisotropy is observed, while when relaxation is taken into account, perpendicular anisotropy predominates.

We have focused on studies concerning the effect of surface relaxation exerted on the value of the energy and magnetic components using the SDFT formalism and calculating the self-consistent band structure. Surface relaxation means a change in the interplanar spacing of atoms, while the shape and volume of the cell remain unchanged.

The effect of ferromagnetic film thickness on the magnetic anisotropy energy has also been studied. The contribution of spin-orbit coupling to the magnetic anisotropy energy has been determined. We have studied all surface faces with a low Miller index.

In our article, we have already studied the effects of anisotropy without taking into account relaxation [12]. The dependence of E_{MA} on the thickness of the substrate and film has been demonstrated. In addition, the anisotropy parameter has been calculated for further applications in Monte Carlo calculations [13]. This article was aimed at studying the effects of anisotropy for a three-layer Fe/Pt/Fe system, as well as the impact of surface relaxation on these effects.

RESULTS AND DISCUSSION

The ab initio calculations were performed using the SDFT implemented with VASP software package [14] based on the method of projected augmented waves (PAW) with the Perdew–Burke–Ernzerhof (PBE) version of the generalized gradient approximation (GGA). The values of the convergence parameters were chosen based on our earlier studies [15].

The article focuses on studies on the relaxation effect on the value of magnetic anisotropy energy E_{MA} and magnetic moments of atoms under varying ferromagnetic Fe film thicknesses, with a fixed thickness of the Pt substrate. We have already studied the effect of the substrate thickness on the E_{MA} value [12].

Here, we emphasized variation of the film thickness from one monolayer to seven atomic layers, with the substrate thickness fixed at three atomic layers. All surface faces with low Miller indices were examined. To conserve the inversion symmetry, the ferromagnetic film was placed on both sides. We obtained the value of the lattice constant earlier from calculations with complete optimization of the crystal structure: 3.87125(3) Å [15].

The energy of magnetic anisotropy was calculated as the difference between two values of the total energy of the system with magnetic moments directed perpendicularly and in parallel to the plane $\Delta E_{MA} = E_{\perp} - E_{\parallel}$. In the case of $\Delta E_{MA} > 0$, the orientation of the magnetic moments parallel to the plane prevails, which corresponds to in-plane anisotropy. In the case of $\Delta E_{MA} < 0$, the orientation of the magnetic moments perpendicular to the plane is energetically more favorable, which corresponds to the easy-axis anisotropy.

For the calculations on the Fe film, the fcc structure of the Pt substrate was used. The use of one atom in the layer in our calculations with the setting of a supercell prevents tracing of the rearrangement transition of the Fe film from the fcc to body-centered cubic (bcc) structure in the case of large film thicknesses. Of course, in E_{MA} studies, it is necessary to take into account structural aspects.

Calculations are required that assume a simultaneous, unlimited change in the geometry of the crystal structure. For such calculations, it is necessary to use several atoms in a layer. However, even one atom in a layer in the case of noncollinear magnetic calculations involves huge computational costs. For example, for a monolayer film, calculations taking into account spin-orbit coupling require 2.5 GB of memory per nucleus and 34 h of calculation time [15].

Relaxation was taken into account as follows. First, surface relaxation of the nonmagnetic system was performed; i.e., only the positions of atoms changed. In this case, the cell size and shape remained unchanged. Then, the obtained relaxed nonmagnetic system was used to study the following two cases. In the first, the magnetic system was calculated with no relaxation taken into account, while conserving the volume, cell shape, and atomic coordinates. In the second case, the the magnetic system was calculated taking into account surface relaxation, i.e., taking into account the change in position of atoms.

Figure 1 shows calculations of the surface relaxation effect on the position of atoms in the system. The surface relaxation parameter δ was calculated, which

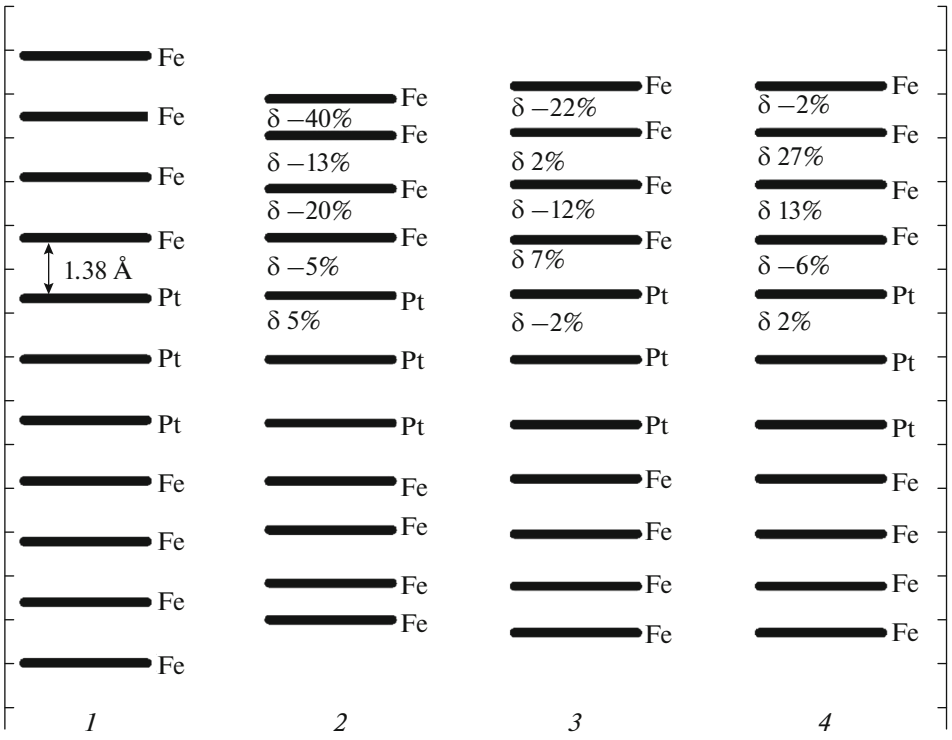

Fig. 1. Effect of surface relaxation on position of atomic planes in Fe/Pt/Fe system; δ, relaxation parameter; (*1*) initial position of atoms; (*2*) nonmagnetic system relaxation; (*3*) relaxation of magnetic system for in-plane direction of magnetization vector; (*4*) relaxation of magnetic system for perpendicular-to-plane direction of magnetization vector.

represents a relative change in the interplanar distance, in percent. Clearly, the interplanar spacing between Fe atoms after relaxation is smaller than that between Pt atoms. The greatest compression was observed near an atom on the surface. The direction of the magnetic moment significantly affects the relaxation parameter, changing not only the value, but also the character of displacement.

Table 1 presents the calculation results for the total energy per atom, depending on the thickness of the ferromagnetic Fe film. Clearly, relaxation exerts an insignificant effect on the reduced energy. The minimum contribution is observed for a monolayer film. With increasing ferromagnetic film thickness, an increase in the relaxation effect on the reduced energy and decrease in energy per atom are observed.

Figures 2a and 2c show the relaxation effect on the average value of magnetization with increasing film thickness. Clearly, with increasing film thickness, the average value of magnetization decreases. In addition, for all the faces, taking relaxation into account leads to an increase in the magnetic moment, except for two cases: for face (111) with a film thickness of three layers and for face (110) with a film thickness of five monoatomic layers. It is just for these systems affected by relaxation that a change in the type of anisotropy is observed, which is shown in Figs. 3a–3c. Figure 3c

Table 1. Calculated values for energy per atom with and without relaxation taken into account for faces (100), (110), and (111)

	(100)		(110)		(111)	
	E, meV/atom.	*E*, meV/atom. with relax.	*E*, meV/atom.	*E*, meV/atom. with relax.	*E*, meV/atom.	*E*, meV/atom. with relax.
Fe_1	−6.782	−6.817	−6.804	−6.830	−6.833	−6.860
Fe_2	−7.163	−7.207	−7.025	−7.060	−7.208	−7.218
Fe_3	−7.344	−7.446	−7.275	−7.329	−7.270	−7.405
Fe_4	−7.498	−7.586	−7.388	−7.446	−7.495	−7.520
Fe_5	−7.611	−7.685	−7.462	−7.685	−7.584	−7.599

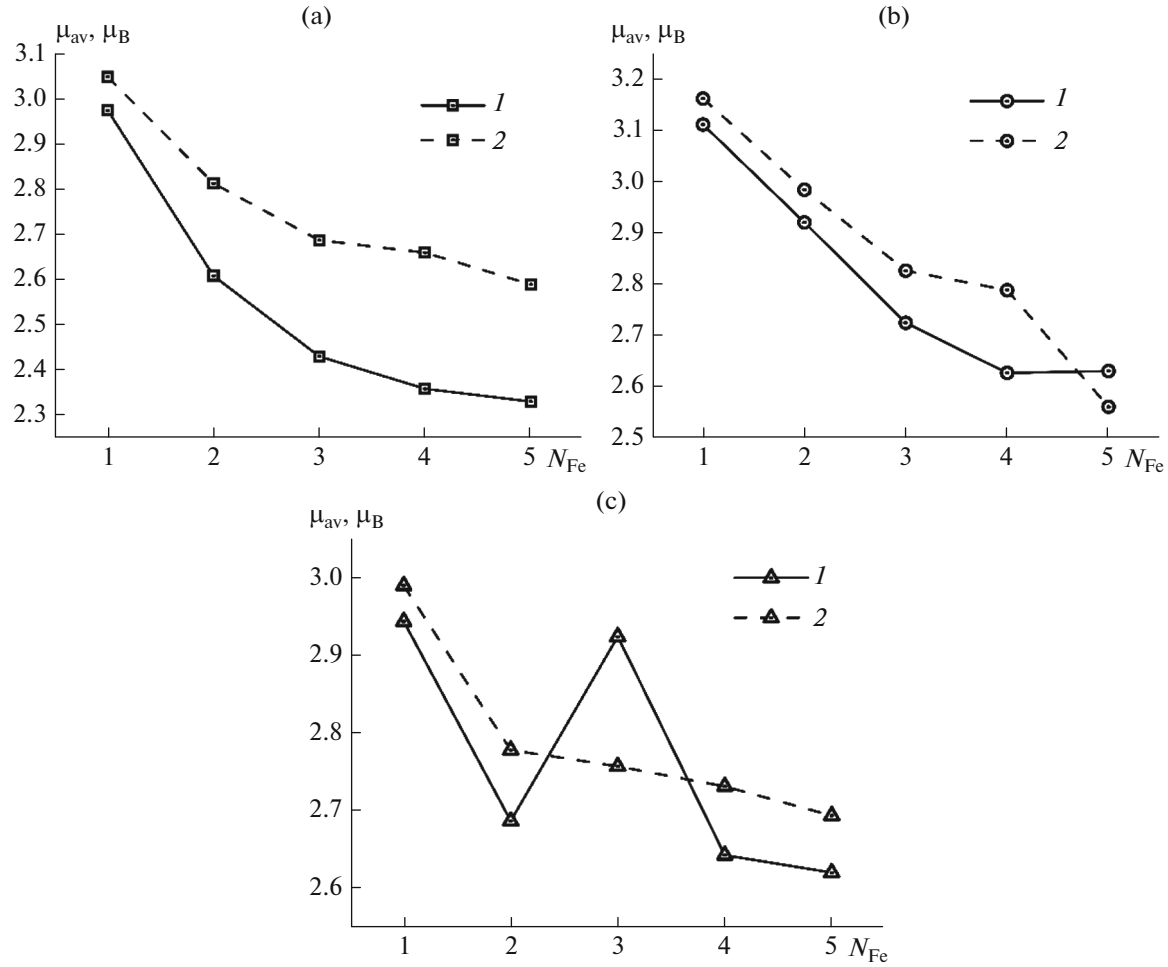

Fig. 2. Value of average magnetic moment depending on thickness of Fe film (a) for (100) faces, (b) for (110) faces, and (c) for (111) faces; without relaxation (*1*) and with relaxation (*2*).

shows a change in the type of anisotropy for a film thickness of three monoatomic layers.

In the calculations without relaxation, in-plane anisotropy predominates, whereas when relaxation is taken into account, easy-axis anisotropy is observed. A similar, although less ample transition can be observed for the (100) face, for a film thickness of five layers (Fig. 3a). In this case, conversely, when relaxation is taken into account, in-plane anisotropy begins to predominate.

For face (111), with increasing film thickness, regardless of the relaxation effect, a transition to predominant perpendicular anisotropy is observed (see Fig. 3c). For faces (100) with increasing film thickness, an oscillatory behavior is characteristic (see Fig. 3a). Each new film layer changes the type of anisotropy from perpendicular to planar and back. For face (110), such strong oscillations like for the (100) face are not observed; however, as the film thickness changes from one to two and three to four monoatomic layers, the type of anisotropy exhibits a change (see Fig. 3b).

The results of our calculations correlate with those for a single Fe monolayer performed by other authors using the Korringa–Kohn–Rostoker method. The authors of [16] obtained the perpendicular magnetic anisotropy for the Pt(100) face. In [11], calculations for the Pt(111) face demonstrate that in-plane magnetic anisotropy with $E_{MA} = 0.71$ meV prevails. Our calculations without relaxation yield $E_{MA} = 2.2$ meV, whereas taking relaxation into account leads to a decrease to $E_{MA} = 1.8$ meV.

The studies on relaxation show that the presence of an inhomogeneous pattern of interatomic distances causes a significant change in the value of E_{MA} and can also be the reason for reorientation of magnetization in atoms. Publications by other authors confirm this for other systems. The authors of [17] have studied the relaxation effect on E_{MA} for the fcc transition metal clusters. In [18], the relaxation effect on E_{MA} for a Co wire at the edge of a Pt(111) step has been studied. In

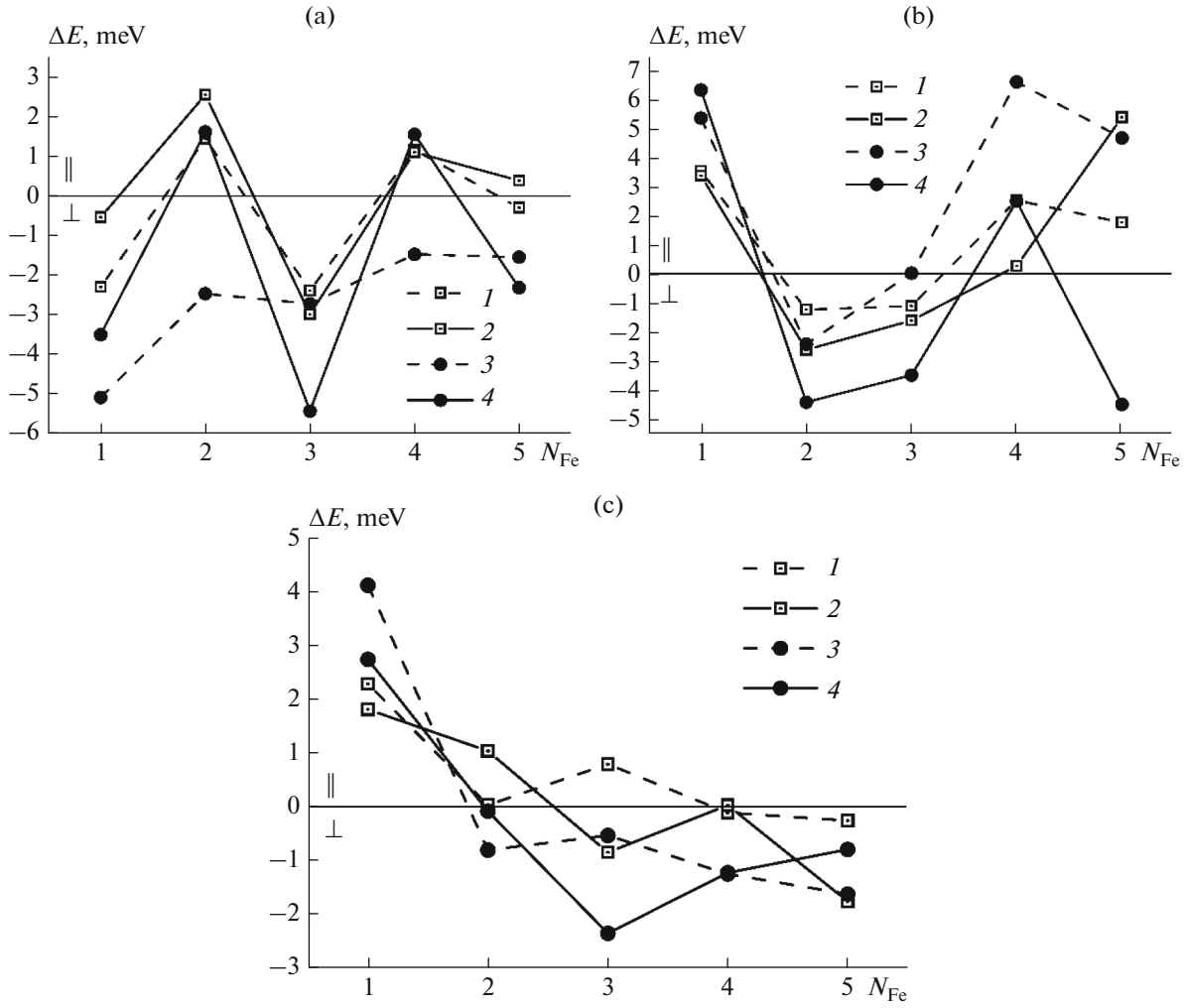

Fig. 3. Value of magnetic anisotropy energy ΔE_{MA} and its spin-orbital component ΔE_{SOC} for Fe/Pt/Fe heterostructure with change in thickness of Fe film: (a) for face (100), (b) for face (110), (c) for face (111); (*1*) ΔE_{MA} without relaxation, (*2*) ΔE_{MA} with relaxation, (*3*) ΔE_{SOC} without relaxation, (*4*) ΔE_{SOC} with relaxation.

both papers, an improvement of the quantitative description of E_{MA} is observed when surface relaxation is taken into account.

When calculating the magnetic anisotropy energy, one should estimate what exactly draws the greatest contribution to the value of E_{MA} i.e., find out where it comes from. In order to answer this question, we have estimated the contribution of the spin-orbit component to the magnitude of the magnetic anisotropy. Inclusion of spin-orbit coupling (SOC) in DFT leads to an additional contribution $H_{SOC}^{\alpha\beta} \propto \vec{\sigma} \cdot \vec{L}$ to the Hamiltonian that relates the Pauli spin operator to the angular momentum $\vec{L} = \vec{r} \times \vec{p}$. As a relativistic correction, the SOC acts predominantly in the immediate vicinity of nuclei, so it is suggested that H_{SOC} contributions beyond the PAW sphere are negligible. Thus, VASP can calculate the matrix elements H_{SOC} only for single-center contributions of all electrons:

$$E_{SOC}^{ij} = \delta_{R_i R_j} \delta_{l_i l_j} \sum_{n,k} w_k f_{nk} \times \sum_{\alpha,\beta} \left\langle \widetilde{\psi_{nk}^{\alpha}} \middle| \widetilde{p_i} \right\rangle \left\langle \phi_i \middle| H_{SOC}^{\alpha\beta} \middle| \phi_j \right\rangle \left\langle \widetilde{p_j} \middle| \widetilde{\psi_{nk}^{\beta}} \right\rangle. \quad (1)$$

The crystal lattice affects the orbitals, which in turn are coupled to spins in the localized case via the spin-orbit coupling. For materials with band gaps, spin-orbit coupling induces an orbital momentum that relates the total magnetic moment to the crystal axes. Table 2 presents the calculated orbital and spin magnetic moments for each Fe atom depending on the film thickness.

From this, one can calculate the total energy that depends on the orientation of the magnetization con-

Table 2. Values of orbital magnetic moment μ_{orb}, μ_B and spin magnetic moment μ, μ_B, for each atom of film depending on film thickness

Atom	$N_{Fe} = 1$, μ_{orb}	$N_{Fe} = 2$, μ	$N_{Fe} = 2$, μ_{orb}	$N_{Fe} = 2$, μ	$N_{Fe} = 3$, μ_{orb}	$N_{Fe} = 3$, μ
Fe_1	0.078	2.943	0.054	2.777	0.097	2.851
Fe_2			0.072	2.779	0.083	2.587
Fe_3					0.087	2.833

Table 3. Calculation results for E_{SOC}, meV, for thickness of magnetic Fe film of three to five monolayers and interlayer thickness of three monolayers, (111) face

	E_{SOC}, meV					
Atom	$N_{Fe} = 5$	$N_{Fe} = 5$ with relax.	$N_{Fe} = 4$	$N_{Fe} = 4$ with relax.	$N_{Fe} = 3$	$N_{Fe} = 3$ with relax.
Pt_1	–749.6	–747.9	–751.1	–747.4	–751.9	–747.8
Pt_2	–752.8	–751.1	–754.1	–750.4	–764.4	–750.5
Pt_3	–749.6	–747.9	–751.1	–747.4	–751.9	–747.8
Fe_1	–9.0	–9.2	–9.1	–9.2	–10.5	–9.1
Fe_2	–11.9	–12.2	–12.1	–12.3	–12.5	–11.7
Fe_3	–12.3	–12.1	–11.4	–11.6	–12.7	–12.9
Fe_4	–11.4	–11.6	–12.4	–12.7		
Fe_5	–12.5	–12.6				

tributed to the anisotropy. Therefore, spin-orbit coupling is the main source of magnetocrystalline anisotropy. In the absence of spin-orbit coupling, the directions of spins have no relationship with the crystal structure, which means that the system is invariant with respect to the overall rotation of all spins. Therefore, all our calculations here take spin-orbit coupling into account.

Figures 3a and 3b, in addition to the magnetic anisotropy energy, show the values of its spin-orbit component for all studied faces. By analyzing the behavior of spin-orbit coupling, it can be concluded that the greatest contribution to the oscillatory behavior of he E_{MA} comes from the energy of spin-orbit coupling.

Pt atoms play an important role in the magnetic anisotropy of the Fe film. The hybridization with Fe orbitals induces spin polarization on the Pt atom and, as a result, increases E_{MA} owing to the large spin-orbit coupling in the Pt atom. This is clearly seen from the calculated values of E_{SOC} for the individual atoms given in Table 3 depending on the magnetic film thickness for the (111) face, and in Table 4, depending on the orientation of the face for a film thickness of five monolayers.

The E_{SOC} value for platinum atoms is almost 70 times greater than that for iron atoms. Except for the Fe atom closest to the interface, the value of E_{SOC} for which the spin-orbit coupling energy is minimal and varies with the thickness to an insignificant extent. The effect of relaxation and face orientation on the E_{SOC} value are comparable in magnitude and do not exceed 10%.

Table 4. Results for E_{SOC}, meV, for thickness of magnetic Fe film of five monolayers for different orientations of surface face taking into account relaxation

Atom	(100)	(110)	(111)
Pt_1	–749.0	–752.5	–747.9
Pt_2	–767.9	–766.8	–751.1
Pt_3	–750.1	–752.4	–747.9
Fe_1	–8.8	–8.6	–9.2
Fe_2	–11.7	–11.7	–12.2
Fe_3	–11.7	–11.8	–12.1
Fe_4	–11.7	–11.7	–11.6
Fe_5	–12.5	–12.5	–12.6

CONCLUSIONS

Thus, the relaxation effect with changing ferromagnetic film thickness on E_{MA} and magnetic moments has been studied. It is shown that the great-

est compression level occurs near the a surface atom, whereas the direction of the magnetic moment significantly affects the relaxation parameter, which changes both the value and character of displacement. Relaxation exerts an insignificant effect on the reduced energy. With increasing ferromagnetic film thickness, a decrease in the value of energy per atom is observed. The relaxation effect increases the values of magnetic moment, except for two cases: for face (111) for a film thickness of three layers and for face (110) for a film thickness of five monoatomic layers, for which a change in the type of anisotropy is observed, affected by relaxation. For the case of the (100) and (110) faces, with increasing film thickness, an oscillating behavior is typical, whereas for the (111) faces, a transition to predominant perpendicular anisotropy is observed. It has been shown that the greatest contribution to the oscillatory behavior of E_{MA} comes from the spin-orbit coupling energy, whereas the value of E_{SOC} for Pt atoms is almost 70 times greater than that for Fe atoms.

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CONFLICT OF INTEREST

The authors declare that they have no conflicts of interest.

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