Non-equilibrium phenomena in magnetic multilayer nanostructures and aging in magnetoresistance

M V Mamonova†, P V Prudnikov‡ and V V Prudnikov§
Dostoevsky Omsk State University, Pr. Mira 55A, Omsk 644077, Russia
E-mail: †mamonova_mv@mail.ru, ‡prudnikovpv@omsu.ru, §prudnikv@mail.ru

Abstract. A Monte Carlo simulation of the non-equilibrium behavior of multilayer magnetic structures Co/Cu(100)/Co and Pt/Co/Cu(100)/Co/Pt characterizing different types of magnetic anisotropy is realized. Simulation of transport properties gives possibility to reveal a nontrivial aging effects in the magnetoresistance of these structures and influence of initial states on two-time dependence of magnetoresistance.

The artificial created multilayer magnetic superlattices has become of great interest for wide range of applications based on the phenomena of the giant magnetoresistance (GMR) and the tunneling magnetoresistance. Devices based on the GMR effect are widely used as read heads of hard disks, memory devices, sensors, etc [1, 2]. The magnetic properties of ultrathin films and superstructures are sensitive to the effects of anisotropy generated by the crystal field of a substrate or nonmagnetic layers. The multilayer magnetic structure Co/Cu(100)/Co extensively usable in active elements of spintronic devices is characterized by anisotropy of "easy" magnetic plane type with magnetization oriented in plane of cobalt film. The structure Pt/Co/Cu/Co/Pt with cobalt films coated by ultrathin platinum films is characterized already by anisotropy of "easy" magnetic axis type with magnetization oriented perpendicularly to plane of cobalt film. As it has been shown in [3], Pt/Co bilayer possess giant energy of magnetic anisotropy and high Curie temperatures attaining 500 K in ultrathin films. Combination of high Curie temperature in cobalt films and perpendicular magnetic anisotropy generated in Pt/Co bilayer makes possible to increase significantly magnetoresistance in Pt/Co/Cu/Co/Pt structure in comparison with Co/Cu/Co structure [4].

The nanoscale periodicity in magnetic multilayer structures gives rise to the mesoscopic effects of the strong spatial spin correlation with the slow relaxation dynamics of magnetization accompanying the quenching of the system in the non-equilibrium state. The experimental investigations of relaxation [5] revealed magnetic aging in a Co/Cr-based magnetic superstructure. We have performed in [6, 7, 8, 9] a numerical Monte Carlo simulation of the non-equilibrium behavior of the multilayer Co/Cr/Co and Co/Cu/Co magnetic structures and revealed the aging effects, which are characterized by slowing down of correlation and relaxation processes with an increase of a waiting time \( t_w \). In contrast to the bulk magnetic systems, where the slow dynamics and aging effects manifest themselves near the critical point [10], the aging in magnetic superstructures is occurred within a wide range of temperatures at \( T \leq T_c \).

The non-equilibrium behavior of a system is realized via its transition at the starting instant \( t_0 \) from the initial state at temperature \( T_0 \) to the state with temperature \( T_s \) differing from \( T_0 \). The evolution of systems with slow dynamics depends on its initial state for times \( t \ll t_{\text{rel}}(T_s) \), where \( t_{\text{rel}}(T_s) \) is a relaxation time at temperature \( T_s \). Various initial states exert noticeable influence
on time dependence of characteristic functions in systems with slow dynamics [10, 11, 12]. In this connection, the non-equilibrium behavior of the system depends on whether it evolves from a high-temperature \( T_0 > T_s \) or a low temperature \( T_0 < T_s \) initial state.

In this paper, a Monte Carlo simulation of the non-equilibrium behavior of multilayer magnetic structures Co/Cu(100)/Co and Pt/Co/Cu(100)/Co/Pt characterizing different types of magnetic anisotropy is carried out. We study manifestation of non-equilibrium behavior of these structures in aging properties of their magnetoresistance. We plan to reveal influence of initial states on two-time dependence of the magnetoresistance in nanostructures with different thicknesses of ferromagnetic films.

We realize in this work a Monte Carlo study of the non-equilibrium behavior of a multilayer magnetic structure (Fig. 1 a) consisting of ferromagnetic films separated by nonmagnetic metal layer.

We consider the array consisting of ferromagnetic films with the thicknesses \( N = 3, 5, 7, 9 \) in units of monatomic layers (ML). The exchange integral \( J_1 \) determining the interaction between the neighboring spins is assumed to be \( J_1/k_BT = 1 \), whereas that for the interlayer interaction is \( J_2 = -0.1J_1 \). The sign of \( J_2 \) is negative because the thickness of nonmagnetic spacers in multilayer structures exhibiting the giant magnetoresistance effect is tuned such that the interlayer exchange interaction effectively provides antiferromagnetism. Owing to this interaction, the magnetization of the neighboring ferromagnetic layers have opposite orientations.

The physical properties of ultrathin films based on Fe, Co, and Ni can be described by the anisotropic Heisenberg model [15, 16] with the Hamiltonian given by the expression

\[
H = - \sum_{<i,j>} J_{ij} [(S_i^x S_j^x + S_i^y S_j^y) + (1 − \Delta_1(N)) S_i^z S_j^z],
\]

(1)

which corresponds to Co/Cu(100)/Co structure with the in plane magnetization. The Hamiltonian in the form

\[
H = - \sum_{<i,j>} J_{ij} [(1 − \Delta_2(N)) (S_i^x S_j^x + S_i^y S_j^y) + S_i^z S_j^z]
\]

(2)
corresponds to Pt/Co/Cu(100)/Co/Pt structure with the out of plane magnetization. Spin \( \vec{S}_i = (S^x_i, S^y_i, S^z_i) \) is determined as the classical unit vector at the \( i \)-th site of a face-centered cubic (fcc) lattice for Co films. \( \Delta_{1,2}(N) \) are parameters characterizing the effective influence of anisotropy generated by the crystal field of the Cu(100) substrate on magnetic properties of Co film subject to its thickness \( N \). The dependence of the anisotropy parameter \( \Delta_{1,2}(N) \) on the film thickness \( N \) is presented in Fig. 1 b.

At the beginning, we calculated the equilibrium characteristics of the multilayer magnetic structure as the magnetization of the films \( \textbf{m}_{1,2} \) with the aim to determine the critical temperatures \( T_c(N) \) of the ferromagnetic phase transition in films with different thicknesses \( N \). For most accurate determination of the critical temperatures we used the method of intersection of curves for temperature dependencies of the Binder cumulant \( U_4(N, T, L) \) for structures with films of different linear sizes \( L = 24, 36, \) and \( 48 \). Metropolis algorithm was used for updating spin configurations. During simulation, \( 10^5 \) MCS/s were discarded for equilibration of spin system, and then measured equilibrium quantities are averaged over \( 10^5 \) MCS/s with 500 runs.

We determined for magnetic structures with film thicknesses \( N = 3, 5, 7, \) and \( 9 \) ML the following values of magnetic ordering temperatures: for structures with anisotropy characterized by the in plane magnetization \( T_c(N = 3) = 2.3108(22), T_c(N = 5) = 2.7342(21), T_c(N = 7) = 2.9072(26), \) and \( T_c(N = 9) = 3.0020(6) \) and for structures with the out of plane magnetization \( T_c(N = 3) = 2.5590(14), T_c(N = 5) = 3.0340(15), T_c(N = 7) = 3.1820(13), \) and \( T_c(N = 9) = 3.2784(15) \).

Simulation of transport properties in Co/Cu/Co and Pt/Co/Cu/Co/Pt structures with current perpendicular to plane (CPP) using methodology \([17, 18]\) have permitted in \([4]\) to calculate temperature dependence of their equilibrium CPP-magnetoresistance values with demonstration that magnetoresistance in Pt/Co/Cu/Co/Pt structures is higher than in Co/Cu/Co structures with the same thickness \( N \). We have used for calculation of the CPP magnetoresistance the two-current Mott model to describe the resistance of different conduction channels \([19]\). It was introduced the resistance of an ferromagnetic film for two groups of electrons with spins up \( R_1 \) and spin down \( R_2 \). As a result, the magnetoresistance of the multilayer structure is determined by the relation:

\[
\delta = \frac{(R_1 - R_2)^2}{4R_1R_2} = \frac{(J_{\uparrow} - J_{\downarrow})^2}{4J_{\uparrow}J_{\downarrow}},
\]

where \( J_{\uparrow,\downarrow} = en_{\uparrow,\downarrow}(V_{\uparrow,\downarrow}) \) is the current density. Here, \( n_{\uparrow,\downarrow} \) is the density of electrons with \( x \) (or \( z \)) components of spin moment equal to \(+1/2\) and \(-1/2\) (axis \( x \) is the quantization axis determined by orientation of magnetization in plane of films for Co/Cu(100)/Co structure and the quantization axis \( z \) for Pt/Co/Cu(100)/Co/Pt structure with out of film plane orientation of the magnetization), \( n = n_\uparrow + n_\downarrow \) is the total electron density and \( \langle V_{\uparrow,\downarrow} \rangle \) are the averaged velocities of electrons with corresponding spin projections. The electron densities with spin up and down can be expressed through the magnetization of the film \( n_{\uparrow,\downarrow}/n = (1 \pm m)/2 \) determined in the process of the Monte Carlo simulation of magnetic properties of the structure. The averaged electron velocity \( \langle V_{\uparrow,\downarrow} \rangle \) can be expressed through the electron mobility and the external electric field intensity \( E \), and after that through the probability of electron displacement in unit time (corresponding to one Monte Carlo step per spin) from unit cell \( i \) to a neighbouring unit cell in the direction of the electric field with averaging over all film unit cells:

\[
\langle V_{\uparrow,\downarrow} \rangle = \mu_{\uparrow,\downarrow}E = \frac{e}{T}E \exp \left( - \frac{\Delta E_{i\uparrow,\downarrow}}{T} \right),
\]

where \( \mu \) is the electron mobility and \( \Delta E_i \) characterizes the change of system energy connected.
with electron jump from \(i\)-cell to a neighbouring cell. \(E_{i,\uparrow,\downarrow}\) is determined by relation
\[
E_{i,\uparrow,\downarrow} = \mp J_1 \left[ \sum_{j \neq i} S_j^z (n_j,\uparrow - n_j,\downarrow) + S_i^z (n_i,\uparrow - n_i,\downarrow) \right]
\]
(5)
in case of magnetization orientation in plane of cobalt films for Co/Cu/Co structure and following relation
\[
E_{i,\uparrow,\downarrow} = \mp J_1 \left[ \sum_{j \neq i} S_j^z (n_j,\uparrow - n_j,\downarrow) + S_i^z (n_i,\uparrow - n_i,\downarrow) \right]
\]
(6)
for Pt/Co/Cu/Co/Pt structure with the magnetization oriented by perpendicularly to plane of ferromagnetic films.

At the next stage of this work, we study of influence of non-equilibrium behavior of the multilayer magnetic structures on their magnetoresistance with realization of evolution from both high-temperature and low-temperature initial states. We calculate two-time dependence of the magnetoresistance \(\delta(t, t_w)\) on observation time \(t - t_w\) and waiting time \(t_w\). The waiting time \(t_w\) characterizes the time between a sample preparation in non-equilibrium initial state and the beginning of measurement of its magnetoresistance.

As an example, we present in Fig. 2 calculated time dependence of the magnetoresistance \(\delta(t, t_w)\) in Co/Cu(100)/Co and Pt/Co/Cu(100)/Co/Pt with the cobalt film thicknesses \(N = 3, 5, 7, 9\) ML on observation time \(t - t_w\) with evolution from the low-temperature completely ordered initial state with \(T_0 = 0\) at temperatures \(T_s = T_c(N)/4\). Values of the magnetoresistance \(\delta(t, t_w)\) were averaged over 250 runs for \(N = 3\) ML and 500 runs for \(N = 7\) ML and 9 ML. The magnetoresistance demonstrates dependence on waiting time \(t_w\) as general criterion of aging and that \(\delta(t, t_w)\) reaches a plateau with asymptotical values \(\delta^\infty(N, T)\), which depend on thickness \(N\) of cobalt films, temperature and type of anisotropy in ferromagnetic films. So, values \(\delta^\infty(N, T)\) are higher for structures Pt/Co/Cu/Co/Pt with easy-axis anisotropy than for structures Co/Cu/Co with easy-plane anisotropy and with the same thickness \(N\) of cobalt films. As can be seen from Fig. 2, difference of values \(\delta^\infty(N, T)\) for Pt/Co/Cu and Co/Cu grows up with increase of cobalt film thickness \(N\). Also, it was revealed that values of \(\delta^\infty(N, T)\) obtained for case of evolution of system from the low-temperature initial state agree very well with equilibrium values of the magnetoresistance \(\delta^{(eq)}(N, T)\).
Figure 3. Time dependence of the magnetoresistance in Co/Cu/Co (a) and Pt/Co/Cu/Co/Pt (b) with different thicknesses $N$ of the cobalt films at temperatures $T_s = T_c(N)/4$ with evolution from the high-temperature initial state with $T_0 \gg T_c(N)$.

We demonstrate in Fig. 3 calculated time dependence of the magnetoresistance $\delta(t, t_w)$ in Co/Cu(100)/Co and Pt/Co/Cu(100)/Co/Pt structures with evolution from the high-temperature completely disordered initial state with $T_0 \gg T_c(N)$ at temperatures $T_s = T_c(N)/4$. Comparison of obtained curves for $\delta(t, t_w)$ in Fig’s. 2 and 3 shows that asymptotical values $\delta^\infty(N, T)$ on plateau for the low-temperature completely ordered initial state are higher than values $\delta^\infty(N, T)$ for case of evolution from the high-temperature completely disordered initial state. Therefore, values $\delta^\infty(N, T)$ for the high-temperature initial state differ from equilibrium values of the magnetoresistance and lower these values. The comparison of $\delta(t, t_w)$ in Fig’s. 2 and 3 shows that the time dependence of magnetoresistance in Co/Cu structures with easy-plane anisotropy reaches a plateau for times about 1 000 - 3 000 MCS/s while in Pt/Co/Cu structures with easy-axis anisotropy for longer times about 3 000 - 6 000 MCS/s.

Figure 4. Time dependence of the magnetoresistance in Pt/Co/Cu/Co/Pt with thickness $N = 5$ ML of the Co films at temperature $T_s = T_c(N = 5)/4 \simeq 241.8$ K with evolution from (a) the intermediary initial states with $T_0^{(ht)} = 3T_c(N = 5)/8 \simeq 362.7$ K and $T_0^{(lt)} = T_c(N = 5)/8 \simeq 120.8$ K and (b) extreme initial states with $T_0^{(ht)} \gg T_c(N = 5)$ and $T_0^{(lt)} = 0$.

Also, we considered influence of intermediary initial states with $0 < T_0 < T_c$ on time dependence of the magnetoresistance. As an example, we present in Fig. 4 calculated $\delta(t, t_w)$ in Pt/Co/Cu(100)/Co/Pt structure with the cobalt film thicknesses $N = 5$ ML with initial temperatures $T_0^{(ht)} = 3T_c(N = 5)/8 \simeq 362.7$ K and $T_0^{(lt)} = T_c(N = 5)/8 \simeq 120.8$ K realized
at quenched temperature $T_q = T_c(N = 5)/4 \approx 241.8$ K. Note that the temperature scale was defined through the value of the exchange integral $J_1 = 4.4 \cdot 10^{-14} \text{erg} \text{ corresponding to the bulk cobalt.}$ This value of $J_1$ is calculated with the use of the well known mean-field approximation. These initial temperatures $T_0^{(ht)}$ and $T_0^{(lt)}$ are the high-temperature and low-temperature states, consequently, in relation to quenched temperature $T_q$. Also, we inserted in Fig. 4 for comparison curves of $\delta(t, t_w)$ for extreme initial temperatures $T_0 \gg T_c(N = 5)$ and $T_0 = 0$. We can see that asymptotical values of the magnetoresistance $\delta^\infty$ on plateau are characterized by sequenced increase of $\delta^\infty$ from values for $T_0^{(ht)} \gg T_c$ and $T_s < T_0^{(ht)} < T_c$ to $0 < T_0^{(lt)} < T_s$ and $T_0^{(lt)} = 0$.

We connect these effects with influence of the effective temperature $T_{\text{eff}} = T/X^\infty$, where $X^\infty$ is the asymptotic value of the fluctuation-dissipation ratio (FDR) [20]. Non-equilibrium critical dynamics of the most statistical model systems is characterized by $X^\infty < 1$ [10]. Values of $X^\infty$ in the multilayer magnetic structures are unknown for temperatures $T_s \leq T_c$, but we can use information about temperature dependence of the FDR with $X^\infty(T) < 1$ and $T_{\text{eff}}(T) > T$ obtained in paper [21] for the 2D XY model. Some community of non-equilibrium properties of the 2D XY model and the multilayer nanostructures permits to declare that $T_{\text{eff}}(T_s) > T_s$ and, consequently, values of the magnetoresistance on plateau $\delta^\infty(N, T_{\text{eff}})$ must be less than equilibrium value of the magnetoresistance for $T_s < T_{\text{eff}}$.

Realized in the present paper Monte Carlo study of the non-equilibrium behavior of Co/Cu(100)/Co and Pt/Co/Cu(100)/Co/Pt nanostructures has revealed nontrivial aging effects in the magnetoresistance $\delta(t, t_w)$ and significant influence of initial states on the magnetoresistance. It has been shown that the magnetoresistance reaches plateau in asymptotical long-time regime with values $\delta^\infty(N, T)$, which depend on type of initial state, thickness of cobalt films, temperature and type of magnetic anisotropy in nanostructures.

Acknowledgments

This work was supported by the Russian Foundation for Basic Research, project No. 20-32-70189, by the Ministry of Education and Science of Russian Federation in the framework of the state assignment No. 0741-2020-0002, and the Council for Grants of the President of the Russian Federation, project No. MD-2229.2020.2.

References

[1] Prinz G A 1999 J. Magn. Magn. Mater. 200 57
[2] Chappert C, Fert A and Van Dau F N 2007 Nature Mater. 6 813
[3] Morgunov R, Hamadeh A, Fache T et al. 2017 Superlattices Microstruct. 104 509
[4] Prudnikov P V, Prudnikov V V, Mamonova M V and Piskunova N I 2019 J. Magn. Magn. Mater. 482 201
[5] Mukherjee T, Pleimling M and Binek C 2010 Phys. Rev. B 82 134425
[6] Prudnikov V V, Prudnikov P V, Mamonova M V 2018 JETP Lett. 104 776
[7] Prudnikov V V, Prudnikov P V and Mamonova M V 2018 JETP 127 731
[8] Prudnikov P V, Prudnikov V V, Purtov A N, Mamonova M V and Piskunova N I 2019 J. Magn. Magn. Mater. 470 143
[9] Prudnikov V V, Prudnikov P V, Mamonova M V, Firstova M M and Samoshilova A A 2019 J. Phys. Comm. 3 015002
[10] Prudnikov V V, Prudnikov P V and Mamonova M V 2017 Phys. Usp. 60 762
[11] Prudnikov V V, Prudnikov P V and Pospelov E A 2016 J. Stat. Mech. 2016 043303
[12] Prudnikov P V, Prudnikov V V, Pospelov E A and Vakilov A N 2015 Phys. Lett. A 379 774
[13] Huang F, Kief M T, Mankey G J and Willis R F 1994 Phys. Rev. B 49 3962
[14] Li Y and Baberschke K 1992 Phys. Rev. Lett. 68 1208
[15] Prudnikov P V, Prudnikov V V and Medvedeva M A 2014 JETP Lett. 100 146
[16] Prudnikov P V, Prudnikov V V, Menshikova M A and Piskunova N I 2015 J. Magn. Magn. Mater. 387 77
[17] Prudnikov V V, Prudnikov P V and Romanovskiy D E 2015 JETP Lett. 102 668
[18] Prudnikov V V, Prudnikov P V and Romanovskiy D E 2016 J. Phys. D: Appl. Phys. 49 235002
[19] Mathon J 1991 Contemporary Phys. 32 143
[20] Cugliandolo L F, Kurchan J and Parisi G 1994 J. Phys. I France 4 1641
[21] Prudnikov V V, Prudnikov P V and Popov I S 2015 JETP Lett. 101 539