Nonequilibrium critical dynamics of low-dimensional frustrated magnets and multilayer structures

P V Prudnikov†, V V Prudnikov, V O Borzilov, M M Firstova, and A A Samoshilova

Department of Theoretical Physics, Omsk State University, Mira prospekt 55-A, Omsk, 644077, Russia
E-mail: †prudnikp@univer.omsk.su

Abstract. A Monte Carlo simulation of the non-equilibrium behavior of multilayer magnetic nanostructure is carried out. Equilibrium properties, critical temperatures \( T_c \) and hysteresis effects were studied for various thicknesses of ferromagnetic layers. It is shown that, in contrast to bulk magnetic systems, the aging effects in nanostructure arise not only at the ferromagnetic ordering temperature \( T_c \) but also within a wide temperature range at \( T \leq T_c \). Simulation of transport properties in multilayer structure permitted to calculate non-equilibrium values of magnetoresistance.

1. Introduction

A significant interest has been recently focused on non-equilibrium processes in magnetic low-dimensional materials. The reduction of the dimension of magnets is accompanied by an increase in fluctuations of the spin density and the manifestation of the effects of critical slowing down and "aging" in the non-equilibrium behavior of low dimensional magnetic systems [1]. Thin films and low-dimensional magnets demonstrate the slow critical evolution from a non-equilibrium initial state. Aging, coarsening and memory effects are nontrivial features in the non-equilibrium behavior of such systems with slow dynamics [2]. The magnetic properties of multilayer magnetic systems have been widely investigated over the past years, since they are widely used in magnetic storage devices [3]. The antiferromagnetic coupling was crucial for the discovery of the giant magnetoresistance (GMR). It kickstarted the field of nanomagnetism and spintronics [4]. The use of synthetic antiferromagnets in magnetic random access memory (MRAM) [5] can reduce the critical current [6] and the time for switching [7].

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. In contrast to the bulk magnetic systems, where the slow dynamics and aging effects manifest themselves near the critical point [2], magnetic superstructures with nanoscale periodicity allow increasing the relaxation time owing to the effects related to the larger characteristic spin-spin correlation length. That is why the aging and nonergodicity effects can be experimentally observed in the multilayer Co/Cr magnetic structure [1] within a wider temperature range as compared to that for the bulk magnetic systems. We have performed in the paper [8] a numerical Monte Carlo simulation of the non-equilibrium behavior of the multilayer Co/Cr magnetic structure.
with the thickness of Co films equals to \( N = 3 \) monolayers. Calculations of the two-time correlation functions and the staggered magnetization allow us to reveal the aging effects, which are characterized by slowing down of correlation and relaxation processes with an increase in the system’s "age" \( t_w \) as the time between a sample preparation and the beginning of measurement of its characteristics. The revealed aging effects for our model of multilayer structure are in good agreement with results of experimental observations in the paper [1]. It was shown that aging in the non-equilibrium critical behavior of multilayer magnetic structure is occurred not only at \( T_c \) but also within a wide range of temperatures at \( T \leq T_c \). Therefore, the existence of these non-equilibrium features should surely be taken into account in any applications of the multilayer magnetic structures for spintronic devices based on the giant magnetoresistance phenomena.

2. Equilibrium properties of the magnetic structure and hysteresis effects

Magnetic order in the multilayers is complex due to a strong influence of the shape and the magnetocrystalline anisotropy of the sample. The presence of anisotropy leads to dimensionality crossover in Heisenberg films [9, 10]. Low-dimensional magnets [11] and multilayers based on anisotropic Heisenberg films [8, 12, 13] demonstrate a dependence on initial states of the non-equilibrium critical evolution. For the statistical Monte Carlo description of ferromagnetic anisotropic Heisenberg films \[9, 10\] demonstrate a dependence on initial states of the non-equilibrium crossover in Heisenberg films \[9, 10\]. Low-dimensional magnets [11] and multilayers based on anisotropic Heisenberg films \[8, 12, 13\] demonstrate a dependence on initial states of the non-equilibrium critical evolution. For the statistical Monte Carlo description of ferromagnetic anisotropic Heisenberg films contacting with nonmagnetic film, we apply the anisotropic Heisenberg model with the Hamiltonian of the spin system in the form

\[
H = -J_1 \sum_{i,j} \left[ S_i \cdot S_j - \Delta(N)S_i^z S_j^z \right] - A \sum_i (S_i^z)^2 - h \sum_i S_i
\]

where \( \vec{S}_i = (S_i^x, S_i^y, S_i^z) \) is a three-dimensional unit vector, fixed in cite \( i \) of the fcc-lattice, \( J_1 > 0 \) is the exchange integral describing the interaction between the neighboring spins in film. The temperature \( T \) of the system is measured in units of the exchange integral \( J_1/k_b \). \( A \) characterizes a single-ion anisotropy. The simulations were performed for films with linear sizes \( L \times L \times N \) with applied periodic boundary conditions in the plane of the film. \( L \times L \) gives the number of spins in each layer, and \( N \) is the number of monolayers in the thin film. \( \Delta(N) \) is the parameter characterizing the effective influence of anisotropy generated by the crystal field of the substrate on magnetic properties of film subject to its thickness \( N \) in terms of monoatomic

![Figure 1](image-url)  

**Figure 1.** Temperature dependence of the staggered susceptibility \( \chi_{stg}(T, N) \) (left) and Binder cumulant \( U_4(T, L) \) for \( N = 3 \). Approximation of cumulant values for different film linear sizes \( L \) is given in insertion. Triangle from crossing lines determines a value of the critical temperature \( T_c \).
layers (ML). $\Delta = 0$ corresponds to the isotropic Heisenberg model, $\Delta = 1$ to the XY-model. The Hamiltonian (1) describes magnetic properties of systems with anisotropy of "easy" magnetic plane type with magnetization oriented in XY plane. The presence of anisotropy leads to the appearance of long-range magnetic order in two-dimensional Heisenberg systems with the critical temperature $T_c \neq 0$.

In this work we consider the multilayer magnetic structure consisting of two ferromagnetic films separated by the nonmagnetic film. The thickness of this nonmagnetic metal layer is selected for obtaining GMR effects in such a way that the long-range and oscillatory RKKY interlayer exchange interaction between spins of the ferromagnetic layers has an effective antiferromagnetic character. Through this interaction, the magnetizations of the adjacent ferromagnetic layers are oriented opposite to each other. When this structure is placed in an external magnetic field, the magnetizations of layers begin to orient parallel that leads to a significant change in the electrical resistance.

We calculated the equilibrium characteristics of the multilayer magnetic structure with the aim to determine the critical temperatures $T_c(N)$ of the ferromagnetic phase transition in films with different thicknesses $N$. For a more accurate determination of the critical temperatures, we considered the structures with films of different linear sizes $L = 20, 32, 40$. We calculated such characteristics as the staggered magnetization $m_{\text{stg}} = m_1 - m_2$, where $m_1$ and $m_2$ are the magnetizations of the films

$$m_n = |m_n| = \frac{1}{N_x} \left[ \sum_{\alpha \in \{x,y,z\}} \left( \sum_{i} S_{i}^{\alpha} \right)^2 \right]^{1/2}, \quad n = 1, 2, \quad (2)$$

the staggered susceptibility (Fig. 1)

$$\chi_{\text{stg}} = \frac{1}{T} \left( \langle m_{\text{stg}}^2 \rangle - \langle m_{\text{stg}} \rangle^2 \right), \quad (3)$$

where $N_x = NL^2$ is the total number of spins in the film and angular brackets denote the statistical averaging.

The most accurate values of the critical temperatures $T_c(N)$ for structures with different film thicknesses $N$ were obtained by the method of intersection of curves for temperature dependencies of the Binder cumulant $U_4(N, T, L)$ for systems with different linear sizes $L$:

$$U_4(N, T, L) = 1/2 \left( 3 - \frac{\langle m_{\text{stg}}^4 \rangle}{\langle m_{\text{stg}}^2 \rangle^2} \right). \quad (4)$$

Figure 2. Magnetic hysteresis loops at $T = 0.8$ for $A = 1$ and $\Delta = 0$: $N_1 = N_2 = 6$ ML symmetric structure (a) $N_1 = N_2 = 6$ ML non-symmetric structure (b)
The scaling dependence of the cumulant

$$U_4(N, T, L) = u(t^{1/\nu}(T - T_c))$$  \hspace{1cm} (5)

makes it possible to determine the critical temperature $T_c(N)$ from the coordinate of the intersection points of the curves specifying the temperature dependence $U_4(N, T, L)$ for different $L$ (Figs. 1). Consequently, we determined for magnetic structures with film thicknesses $N = 3, 5, 7, 9$ ML the following values of magnetic ordering temperatures: $T_c(N = 3) = 2.5413(8)$, $T_c(N = 5) = 2.9033(12)$, $T_c(N = 7) = 3.0356(6)$, $T_c(N = 9) = 3.1014(11)$.

The magnetic hysteresis loops for the multilayer system at $T = 0.8$ with $A = 1$ and $\Delta = 0$ are presented in Figure 2. It is interesting to see that the hysteresis loops are changed with increasing of exchange interlayer interaction. The presence of new additional steps on hysteresis loop is connected with additional magnetic states (Figure 2a). The shape of the hysteresis loop for non-symmetric structure (Figure 2b) is formed by three subsequent transitions between ferro-antiferro-ferromagnetic states.

3. Non-equilibrium behavior of the magnetic structure and aging effects

As it was shown in [1,8], the effects of slow dynamics appear in magnetic superstructures within a wide range of temperatures at $T \leq T_c$.

One of important features arisen in non-equilibrium behavior of systems with slow dynamics [2] is dependence on an initial states. The non-equilibrium behavior of a system is realized during 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 accompanying equilibration process is characterized by relaxation time $t_{rel}(T_s)$, and equilibrium corresponding to temperature $T_s$ is reached in times $t \gg t_{rel}(T_s)$, while the system dynamics prove stationary and invariant with respect to time reversal. However, in times $t \ll t_{rel}(T_s)$, the evolution of the system depends on its initial state. 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. The high-temperature initial state for magnetic systems is created at $T_0 > T_c$ and characterized by initial magnetization $m_0 = 0$, while the low-temperature initial states with $T_0 < T_s$ are characterized by $m_0 \neq 0$. The non-equilibrium behavior of a systems with slow-dynamics demonstrates the breakdown of translational invariance in time due to the long-time influence of non-equilibrium initial states. It manifests itself through two-time characteristics of the system, such as the autocorrelation function

$$C(t, t_w) = \frac{1}{V} \int d^d x [\langle S(x, t) S(0, t_w) \rangle - \langle S(x, t) \rangle \langle S(0, t_w) \rangle],$$  \hspace{1cm} (6)

where the waiting time $t_w$ is the time between a sample preparation and the beginning of measurement of its characteristics and $t - t_w$ is the time of observation with $t - t_w, t_w \ll t_{rel}$.

Description of the non-equilibrium critical behavior of a number of model statistical systems such as three-dimensional Ising model, the two-dimensional XY model, and multilayer magnetic structure [2] shows that the two-time dependent quantities demonstrate so-called aging. This phenomenon is characterized by both translation symmetry breaking in time and a slowdown of relaxation and correlation processes with increasing ”age” $t_w$ of the sample. The aging is displayed most pronouncedly in two-time dependence of the autocorrelation function (6) during evolution of system from the high-temperature initial state. The aging effects in investigated magnetic structure dependence on ferromagnetic film thicknesses $N$ in a wide quenching temperature range $T_b \leq T_c$, we concentrate our efforts uppermost on study of autocorrelation function two-time behavior with evolution from the high-temperature initial state with $T_0 > T_c$ and $m_0 = 0$. 

4
Figure 3. Autocorrelation function versus the observation time $t - t_w$ for structure with films thickness $N = 5$ ML at quenching temperatures $T_s = T_c = 2.9033$, $(5/6)T_c$, and $(1/2)T_c$.

We computed the two time-dependent autocorrelation function taken in the form

$$C(t, t_w) = \left< \frac{1}{N_s} \sum_{i=1}^{N_s} S_i(t) S_i(t_w) \right> - \left< \frac{1}{N_s} \sum_{i=1}^{N_s} S_i(t) \right> \left< \frac{1}{N_s} \sum_{i=1}^{N_s} S_i(t_w) \right>.$$  \hspace{1cm} (7)

The simulated structures were characterized by the linear size of the films $L = 64$ and thicknesses $N = 3, 5, 7, 9$ ML. The quenching temperatures $T_s(N)$ were taken equal and also fractional to critical temperatures, i.e., $T_s = T_c(N)$, $(5/6)T_c(N)$, $(2/3)T_c(N)$, and $T_c(N)/2$. We used the waiting times $t_w = 10, 30, 50, 100$ MCS/s in the study of the two-time dependence of $C(t, t_w)$. Statistical averaging of the autocorrelation function $C(t, t_w)$ was carried out on 500 MC runs for every $t_w$.

As an example, the obtained time dependence of the autocorrelation function $C(t, t_w)$ from observation time $t - t_w$ for waiting time values $t_w = 30, 50, 100$ MCS/s is presented in Fig. 3 for structure with films thickness $N = 5$ ML at quenching temperatures $T_s = T_c = 2.9033$, $(5/6)T_c$, and $(1/2)T_c$.

The curves of $C(t, t_w)$ clearly demonstrate the aging effects in this magnetic structure, i.e., the slowing down of time correlations with increasing system age $t_w$. We must note that the aging in the multilayer structures arises not only at $T_s = T_c$ as in bulk systems, but also in the low-temperature phase at $T_s < T_c$. Furthermore, the comparison of curves in Fig. 3 at temperatures $T_s = T_c$, $(5/6)T_c$, and $(1/2)T_c$ shows that the aging effects become stronger with decrease of quenching temperature relative to the critical temperature, i.e., the falling of time correlations at $T_s < T_c$ with the same waiting times $t_w$ become more slow than at the critical temperature. Reason for this behavior is connected with XY-type anisotropy, which is realized in Co/Cu(100)/Co structures, and with extremely slow dynamics in two-dimensional XY-model characterized by aging not only near the temperature of the Berezinskii-Kosterlitz-Thouless phase transition $T_{BKT}$, but also in the entire range of the existence of the low-temperature phase [15–18].

At the next stage of this work, we study of influence of non-equilibrium behavior of the multilayer magnetic structure on its magnetoresistance with realization of evolution from both high-temperature and low-temperature initial states with the films magnetization $m_0 = 0$ and $m_0 = 1$, correspondingly. For magnetoresistance calculations we used methods, which were developed in [19, 20].

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
Figure 4. Time dependence of the magnetoresistance $\delta$ for the $N = 5$ at temperatures $T_s = T_c(N)/4$ for different waiting times $t_w = 100, 200, 400,$ and $1000$ MCS/s with evolution from (a) the high-temperature and (b) low-temperature initial states.

Figure 4 shows the time dependence of the magnetoresistance $\delta$ for the $N = 5$ at temperatures $T_s = T_c(N)/4$ for different waiting times $t_w = 100, 200, 400,$ and $1000$ MCS/s with evolution from the high-temperature initial state (Fig. 4a) and the low-temperature initial state (Fig. 4b) at temperatures $T_s = T_c(N)/4$ (for $N = 5$, $T_s \approx 231.5$ K). Values of the magnetoresistance $\delta(t, t_w)$ given in Fig. 4 were averaged over 250 runs.

4. Conclusions
In the present paper a Monte Carlo simulation of the non-equilibrium behavior of multilayer nanostructure is carried out with consideration of different thicknesses $N$ of the ferromagnetic films and variation of temperature in wide range with $T \leq T_c(N)$. Two-time dependencies of the autocorrelation function demonstrate aging effects in magnetic structure for all temperatures in low-temperature phase. This phenomenon is characterized by slowing-down of correlation processes with increasing the waiting time $t_w$, and it is connected with increasing correlation length for transverse spin-spin correlations when the temperature is decreased that leads to increasing correlation and relaxation times in these systems.

We have realized after that the study of influence of non-equilibrium behavior in Co/Cu(100)/Co structure on its magnetoresistance. It is revealed aging effects in time dependence of the magnetoresistance $\delta(t, t_w)$, which is characterized by dependence of its values on waiting time $t_w$ as the beginning of magnetoresistance measurement. The existence of these non-equilibrium effects should surely be taken into account in any applications of the multilayer magnetic structures for spintronic devices based on the giant magnetoresistance effect.

Acknowledgments
This research was supported by the grants 17-02-00279, 18-42-55003 of Russian Foundation of Basic Research and by the grant MD-6868.2018.2 of the Council of the President of the Russian Federation.

The simulations were supported in through computational resources provided by the Shared Facility Center “Data Center of FEB RAS” (Khabarovsk), by the Supercomputing Center of Lomonosov Moscow State University, by Moscow Joint Supercomputer Center and by St.
Petersburg Supercomputer Center of the Russian Academy of Sciences.

References

[1] Mukherjee T, Pleimling M, and Binek Ch 2010 Phys. Rev. B 82 134425
[2] Prudnikov V V, Prudnikov P V, and Mamonova M V 2017 Phys. Usp. 60 762
[3] Chen Y, Song D, Qiu J, et al. 2010 IEEE Trans. Magn. 46 697
[4] Duine R A, Lee K-J, Parkin S S P, Stiles M D 2017 arxiv.org:1705.10526
[5] Apalkov D, Diony B, Slaughter J M 2016 Proceedings of the IEEE 104 1796
[6] Hayakawa J, Ikeda S, Lee Y M, Sasaki R, Meguro T, Matsukura F, Takahashi H, and Ohno H 2006 Jpn. J. Appl. Phys. 45 L1057
[7] Bergman A, Skubic B, Hellsvik J, Nordström L, Delin A, and Eriksson O 2011 Phys. Rev. B 83 224429
[8] Prudnikov V V, Prudnikov P V, Purtov A N, Mamonova M V 2016 JETP Lett. 104 776
[9] Prudnikov P V, Prudnikov V V, Menshikova M A, and Piskunova N I 2015 J. Magn. Magn. Mater. 387 77
[10] Prudnikov P V, Prudnikov V V, Medvedeva M A 2014 JETP Lett. 100 446
[11] Popov I S, Prudnikov P V, Ignatenko A S, Katanin A A 2017 Phys. Rev. B 95 134437
[12] Prudnikov V V, Prudnikov P V, Purtov A N, Mamonova M V, Piskunova N I 2018 J. Magn. Magn. Mater. doi:10.1016/j.jmmm.2017.11.084
[13] Prudnikov V V, Prudnikov P V, Mamonova M V 2018 JETP 127 731
[14] Huang F, Kief M T, Mankey G J, and Willis R F 1994 Phys. Rev. B 49 3962
[15] Berthier L, Holdsworth P C W, Sellitto M 2001 J. Phys. A 34 1805
[16] Prudnikov V V, Prudnikov P V, Alekseev S V, Popov I S 2014 Phys. Met. Metall. 115 1186
[17] Prudnikov V V, Prudnikov P V, Popov I S 2015 JETP Lett. 101 539
[18] Prudnikov V V, Prudnikov P V, Popov I S 2018 JETP 126 369
[19] Prudnikov V V, Prudnikov P V, and Romanovskiy D E 2016 J. Phys. D: Appl. Phys. 49 235002
[20] Prudnikov V V, Prudnikov P V, Romanovskii D E 2015 JETP Lett. 102 668