Non-equilibrium critical behavior of multilayer magnetic nanostructures with a non-magnetic metal substrate

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Abstract. A Monte Carlo simulation of the non-equilibrium behavior of multilayer magnetic nanostructure Co/Cu(100)/Co is realized. Calculations of two-time dependent autocorrelation function for structures relaxing from both low-temperature and high-temperature initial states reveal occurrence of aging within a wide range of temperatures at $T \leq T_c$.

1. Introduction

The physics of ultrathin magnetic films and multilayer nanostructures on their basis is characterized by intensive research over the past decades [1]. This heightened interest of scientists is caused by quite many unique properties of these films considerably different from properties of bulk materials. The use of ferromagnetic films containing Co in artificial created multilayer superlattices leads to interesting phenomena, such as the effect of giant magnetoresistance [2, 3].

The nature of magnetic order in ultrathin ferromagnetic films is very complex due to the competition between exchange and dipolar interactions on different length scales, together with a strong influence of the shape and the magnetocrystalline anisotropy of the sample. These, in turn, are very susceptible to the growth conditions of the films. In the past years, a considerable amount of experimental results on different aspects of magnetism in ultrathin films has been appeared [3, 4]. In this paper, we review some key concepts in ultrathin film magnetism and simulation of equilibrium properties of the multilayer structure. Ultrathin film magnetism has long history since matured into an active area of research, with important technological ramifications. Surely, thin film magnetism has brought important contributions to our fundamental understanding of the physics of magnetism in tandem with critical applications in information technology, particularly evident since the discovery of the giant magnetoresistance effect and following its impact on computer read-head technology.

We have carried out in paper [5] a Monte Carlo simulation of the non-equilibrium behavior of the multilayer $Co/Cr/Co$ structure with different thicknesses of Co films. Calculations of the staggered magnetization and the two-time correlation functions allowed 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 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 the results of experimental observations in [6]. The existence of these non-equilibrium features should surely be taken into account in any applications of the multilayer ultrathin magnetic structures for spintronic devices such as memory hard disks based on the giant magnetoresistance phenomena.

In this work, we present a Monte Carlo study of the non-equilibrium behavior of multilayer Co/Cu(100)/Co nanostructure, used widely in active elements of spintronics devices, with different thicknesses \( N \) of cobalt films and within a broad range of temperatures at \( T \leq T_c(N) \).

2. Modeling of equilibrium properties of the multilayer Co/Cu/Co structure

We have observed in [7, 8] thickness-dependent magnetic properties of ultrathin multilayer films Co, Ni, and Fe on nonmagnetic metal substrates which are determined generally by interfacial interaction films with a substrate and magnetic anisotropy generated by the crystal field of the substrate as well as their changes with increasing of thicknesses \( N \) of the magnetic films.

For description of ferromagnetic Co films contacting with Cu film with surface plane orientation (100), we apply the anisotropic Heisenberg model with the Hamiltonian of the spin system in the form

\[
H = -J_1 \sum [S_i S_j - \Delta(N) S^z_i S^z_j],
\]

where \( J_1 > 0 \) is the exchange integral describing the interaction between the neighboring spins in film, \( S_i = S_i^x, S_i^y, S_i^z \) is a three-dimensional unit vector, fixed in site \( i \) of the Co fcc-lattice. \( \Delta(N) \) in (1) is the parameter characterizing the effective influence of anisotropy generated by the crystal field of the substrate on magnetic. \( \Delta(N) = 0 \) corresponds to the isotropic Heisenberg model and \( \Delta(N) = 1 \) to the XY-model [9].

Dependence of the anisotropy parameter \( \Delta(N) \) on thickness \( N \) Co film is described by a decrease with a thickness \( N \) growth because of the dimensional dependence of the magnetic characteristics shows the transition from the two-dimensional values in films with thickness less than 3 - 5 monolayers to the bulk valuers for films with thicknesses of several tens monolayers, which is accompanied by the weakening of anisotropy impact and demonstration of isotropic magnetic properties [10]. The values of the anisotropy parameter \( \Delta(N) \) presented in Table 1 were calculated on basis of experimental data [11] for the critical temperatures \( T_c \) in films Co/Cu(100) as quantity determined by the relative change of the ferromagnetic transition temperature \( T_c(N) \) in films of Co to \( T_c(\infty) \) in the bulk sample of cobalt.

We consider the multilayer magnetic structure consisting of ferromagnetic Co films separated by nonmagnetic Cu layer. The Hamiltonian for simulation of this structure is taken in the form

\[
H = -J_1 \sum [S_i S_j - \Delta(N) S^z_i S^z_j] - J_2 \sum [S_i S_j - \Delta(N) S^z_i S^z_j],
\]

where the exchange integral \( J_1 > 0 \) defines the intralayer interaction, whereas \( J_2/J_1 = -0.3 \) determines the interlayer interaction. The simulations were performed for ferromagnetic films with linear sizes \( L \times L \times N \) with applied periodic boundary conditions in the plane of the film. \( N \) gives the number of monolayers in the thin Co film.

Values of critical temperature \( T_c(N) \) (Table 1) were calculated by Binder’s cumulants method. For updating spin configurations, we have used the Metropolis algorithm, which generates correct single-spin flip relaxation dynamics. During the simulation of equilibrium properties, \( 10^5 \) MCS/s were performed for the equilibrium of the spin system and then measured equilibrium quantities are averaged over \( 10^5 \) MCS/s with 300 runs.

\[2\]
Table 1. Temperatures for the study of various film thicknesses and anisotropy parameter [8]

| N | $T_c$ | $1/2T_c$ | $1/4T_c$ | $\Delta(N)$ |
|---|---|---|---|---|
| 3 | 2.54 | 1.27 | 0.64 | 0.66 |
| 5 | 2.90 | 1.45 | 0.73 | 0.48 |
| 7 | 3.04 | 1.52 | 0.76 | 0.34 |
| 9 | 3.10 | 1.55 | 0.77 | 0.25 |

3. Non-equilibrium behavior of Co/Cu/Co structure with evolution from high-temperature initial state

Great feature arisen in the non-equilibrium behavior of systems with slow dynamics [12] is dependence on an initial state. The non-equilibrium behavior of a system is realized during the 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 non-equilibrium behavior of a system with slow dynamics demonstrates the breakdown of translational invariance in time due to the long-time influence of non-equilibrium initial states [12]. It manifests itself through two-time characteristics of the system, such as the autocorrelation functions

$$C(t, t_w) = \frac{1}{V} \int d^d x \left[ \langle S(x, t)S(x, t_w) \rangle - \langle S(x, t) \rangle \langle S(x, t_w) \rangle \right]$$  (3)

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

Description of the non-equilibrium critical behavior of many model statistical systems such as the two-dimensional XY-model, the three-dimensional Ising model, and the multilayer magnetic structure Co/Cr/Co given in review [12] shows that the two-time dependent quantities demonstrate so-called aging effects.

In present work, we calculated time dependence of the autocorrelation function $C(t, t_w)$ for waiting time values $t_w = 30$ and 100 MCS/s which is presented in Fig’s. 1-4 for structures with Co films thickness $N = 3 \div 9$ ML at quenching temperatures $T_s(N)$, $1/2T_c(N)$ and $1/4T_c(N)$ for evolution structure from the high-temperature initial state with $T_0 \gg T_c(N)$.

The curves of $C(t, t_w)$ in Fig’s. 1-4 clearly demonstrate the aging effects in this magnetic structure, i.e., the non-exponential slowing down of time correlations with increasing system age $t_w$. We can see that aging in the non-equilibrium critical behavior of a multilayer magnetic structure is occurred not only at $T_c$ but also within a wide range of temperatures at $T \leq T_c(N)$.

The comparison of curves in Fig’s. 1-4 at temperatures $T_s(N) = T_c(N)$, $1/2T_c(N)$ and $1/4T_c(N)$ shows that the aging effects becomes stronger with decrease of quenching temperature $T_s(N)$ relative to the critical temperature $T_c(N)$, i.e. the falling of time correlations at $T_s < T_c$ with the same waiting times $t_w$ becomes 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 [9].

For the aging regime at $t \sim t_w$, the two-time $C(t, t_w)$ has the scaling form

$$C(t, t_w) \sim t_w^{-b} F_c(t/t_w)$$  (4)

where the exponent $b$ at the temperature $T_s = T_c$ is expressed in terms of the critical exponents as $b = 2\beta/\nu z$. The scaling function $F_c(t/t_w)$ is a homogeneous function of its argument.
Figure 1. The time dependence of the autocorrelation function $C(t,t_w)$ for waiting times $t_w = 30$ and 100 MCS/s for structure with $N = 3$ at temperatures $T_c$, $1/2T_c$, and $1/4T_c$ with evolution from high-temperature initial state.

Figure 2. The time dependence of the autocorrelation function $C(t,t_w)$ for waiting times $t_w = 30$ and 100 MCS/s for structure with $N = 5$ at temperatures $T_c$, $1/2T_c$, and $1/4T_c$ with evolution from high-temperature initial state.

Figure 3. The time dependence of the autocorrelation function $C(t,t_w)$ for waiting times $t_w = 30$ and 100 MCS/s for structure with $N = 7$ at temperatures $T_c$, $1/2T_c$, and $1/4T_c$ with evolution from high-temperature initial state.

Figure 4. The time dependence of the autocorrelation function $C(t,t_w)$ for waiting times $t_w = 30$ and 100 MCS/s for structure with $N = 9$ at temperatures $T_c$, $1/2T_c$, and $1/4T_c$ with evolution from high-temperature initial state.

t/t_w. Estimated values of calculated critical exponents are $b(T_c,N = 3) = 2\beta/\nu z = 0.318$, $b(T_c,N = 5) = 0.414$, $b(1/2T_c,N = 3) = 0.05$, $b(1/2T_c,N = 5) = 0.09$. These values correspond to universality class of 2D XY-model [9].

4. Non-equilibrium behavior of Co/Cu/Co structure with evolution from low-temperature initial state
In the work, we got time dependence of the autocorrelation function $C(t,t_w)$ for waiting time values $t_w = 30$ and 100 MCS/s which is presented in Fig’s. 5 and 6 for structures with Co
films thickness $N = 3$ ML and $N = 5$ ML, consequently, at quenching temperatures $T_s(N)$ and $1/2T_c(N)$. We study the systems with evolution from the low-temperature initial state with $T_0 = 0$.

The curves of autocorrelation functions $C(t, t_w)$ in this case also demonstrate the aging effects, i.e., the non-exponential slowing down of correlation processes with increasing the waiting time $t_w$. Comparison of $C(t, t_w)$ curves in Fig’s. 1 and 2 with curves in Fig’s. 5 and 6 shows that correlation times for case of systems evolution from a high-temperature initial state are much greater than those in the evolution from a low-temperature initial state at the same $t_w$ values. For case of evolution from the low-temperature initial state, it is observed that correlation times are two-three orders of magnitude smaller than those in the evolution from a high-temperature initial state at the same $t_w$ values. This time behavior of the autocorrelation function doesn’t depend considerably on temperature for $T_s \leq T_c$ and thickness $N$ of cobalt films.

5. Conclusions
In the present work, we provided a Monte Carlo simulation of the non-equilibrium behavior of multilayer nanostructure Co/Cu/Co, using extensively in the active elements of spintronics devices. Research is carried out with consideration of thicknesses $N = 3 \div 9$ ML of the Co films and variation temperature in a broad range with $T \leq T_c(N)$.

Two-time dependence of the autocorrelation function, determined for both cases of system evolution from low-temperature and high-temperature initial states, demonstrate aging effects in magnetic structure for all temperatures in the low-temperature region. Aging effects are characterized by the slowing-down of correlation processes with increasing the waiting time $t_w$. For the event of the high-temperature initial state, the strengthening of aging effects is revealed with the decrease of quenching temperature $T_s$ relative to the critical temperature $T_c$, which is characterized by greater slowing-down of correlation processes with increasing $t_w$ for lower temperatures $T_s < T_c(N)$. For the event of the low-temperature initial state, it is shown that correlation times are two-three orders of magnitude smaller than those in the evolution from a high-temperature initial state at the same $t_w$ values. In this case, the time behavior of
the autocorrelation function doesn’t depend considerably on temperature for $T_s < T_c(N)$ and thickness $N$ of Co films.

The results show the presence of aging effects in the system, i.e. time dependencies of correlation effects depending on latency of $t_w$. Analysis of the graphs shows that the effects of aging occur in multilayer structures, not only at a critical temperature but also in a wide low-temperature region. 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.

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References

[1] Vaz C A F, Bland J A C and Lauhoff G 2008 *Rep. Prog. Phys.* 71 056501
[2] Chappert C, Fert A and Van Dau F N 2007 *Nature Mater.* 6 813
[3] Bland J A C and Heinrich B 2005 *Ultrathin Magnetic Structures IV* (Springer, Berlin)
[4] Portmann O 2006 *Micromagnetism in the Ultrathin Limit* (Logos Verlag, Berlin)
[5] Prudnikov V V, Prudnikov P V, Purto A N and Mamova M V 2016 *JETP Lett.* 104 776
[6] Mukherjee T, Pleimling M and Binek C 2010 *Phys. Rev.* B 82 134425
[7] Prudnikov P V, Prudnikov V V and Medvedeva M A 2014 *JETP Lett.* 100 446
[8] Prudnikov P V, Prudnikov V V, Menshikova M A and Piskunova N I 2015 *J Magn. Magn. Mater.* 387 77
[9] Prudnikov V V, Prudnikov P V and Popov I S 2015 *JETP Lett.* 101 539
[10] Prudnikov V V, Prudnikov P V, Mamova M V, Samoshilova A A and Firstova M M 2019 *J. Phys. Commun.* 3 015002
[11] Huang F, Kief M T, Mankey G J and Willis R F 1994 *Phys. Rev.* B 49 3962
[12] Prudnikov P V, Prudnikov V V and Mamova M V 2017 *Phys. Usp.* 60 762