Writing the regular domain structures in ultra-thin ferromagnetic films by short trains of ultra-short laser pulses

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ABSTRACT

A new laser technology, based on spatially non-uniform illumination of ultra-thin ferromagnetic films by short trains of ultra-short laser pulses and enabling to create the regular domain structures, is presented.

Keywords: Ultra-thin ferromagnetic films, ultra-short laser pulses, spatially, non-uniform illumination, remagnetization rate, regular domain structures

1. INTRODUCTION

Interest in processes proceeding in ultra-thin ferromagnetic films upon their optical excitation has quickened in the last years due to a rapid development of micro- and nano-technologies, in particular, due to the necessity of increasing the density and rate of magnetic data recording [1]. To solve these purely applied problems, the ultra-fast magnetization kinetics should be studied in detail. The analysis of this kinetics cannot be restricted to the traditional consideration of electron-electron (e-e), electron-phonon (e-ph), and spin-phonon (s-ph) scattering, and along with these processes one should also take into account the spin-orbit and exchange interactions, scattering of carriers by surfaces, etc. [2]. The consideration of a rather complicated real band structure of transition metals, of the possibility of the interaction between electrons from different bands, of the movement of domain walls, and other processes makes this analysis even more involved [3,4].

Among modern experimental techniques used for studying the magnetization kinetics, optical methods are most efficient. There exist several different opinions in the literature about the demagnetization rate of ultra-thin ferromagnetic films. Thus, measurements of the kinetics of transmission of probe light pulses by initially ferromagnetic films after their optical excitation have shown that the spin relaxation time is ~ 1 ps [5]. At the same time, experiments on the second harmonic generation from surfaces of such films have not revealed a noticeable difference between the relaxation rates of the electron temperature and spin [6]. In fact, this means that magnetization should relax during the time ~ 100 fs. Two-photon emission spectroscopy experiments have shown the presence of several demagnetization processes with characteristic times in the range from 10^{-12} to 10^{-10} s [7]. "Direct" Kerr-effect measurements of the magnetization [8] have demonstrated that the result of the experiment depends on the pump-pulse duration τ_p. Upon excitation of ferromagnetic films by rather long laser pulses ($\tau_p > 10$ ns), when the electron temperature T_e should not exceed the Curie temperature T_K, the magnetization was completely destroyed. However, when 20-ps pump pulses were used, no magnetization destruction was observed, although in this case the electron temperature T_e should be much higher than T_K. The results of picosecond ($\tau_p = 20$ ps) nonlinear spectroscopy of ultra-thin (~17-nm thick) Ni films by the biharmonic pumping (BP) technique [9] and of the degenerate four-photon spectroscopy (DFPS) [10] also are contradictory. While the DFPS experiments showed that the demagnetization time in such films should at least exceed 20 ps, the BP measurements gave the opposite result.

In this paper, we will attempt to explain why the results of two spectroscopic experiments [9,10], which have been performed under the same conditions by two methods of picosecond four-photon spectroscopy, contradict to each other. We propose a model that takes into account all main processes proceeding under the experimental conditions [9,10] (during and after the action of picosecond optical pulses on the domain structure of ferromagnetic films). Our numerical simulations show that the demagnetization (remagnetization) rates of ferromagnetic films in BP experiments upon spatially uniform excitation and in DFPS experiments upon spatially non-uniform excitation should be substantially

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different. A new laser technology, based on spatially non-uniform illumination of ultra-thin ferromagnetic films by short trains of ultra-short laser pulses and enabling to create the regular domain structures, will be presented.

2. THE MODEL

We will assume that the total spin of the system is conserved at the time scale of τ_p. This assumption is based on the results obtained in a number of papers. The authors of paper [2] performed a rather realistic numerical calculation (taking into account the real band structure, exchange processes, spin-orbit interaction, etc.) of the spin relaxation time τ_s of excited electrons. They showed that τ_s strongly depends on the excess energy E of free carriers (measured from the Fermi level E_F) and $\tau_s \sim 100$ fs for $E - E_F \sim 2$ eV. Under the experimental conditions [9,10] ($\tau_p = 20$ ps) of our interest, the excited carriers are rapidly thermalized ($\tau_{e-e} \sim 280$ fs [6]) despite a comparatively high (~ 2 eV) photon energy, and although the electron temperature T_e differs from the phonon temperature T_{ph}, we have $T_e \leq 1000$ K and $E - E_F \leq 85$ meV. Because for $0 \leq E - E_F << E_F$ the rate of relaxation processes in a Fermi liquid depends on E as $\tau^{-1} = (2\pi\hbar)\rho^3 |M|^2 (E - E_F)^2$ [3], for $E - E_F = 85$ meV, taking into account the data [2] (see above), the estimate of τ_s gives ~55 ps. Here ρ is the density of the electronic states near the Fermi surface and M is the transition matrix element. Both these parameters were assumed constant in estimating τ_s. Thus, based on the experiments [8], whose conditions approximately correspond to the experimental conditions [9,10], and on paper [11], we can really assume in our calculations that the total spin of the system is conserved at the time scale of τ_p.

We will describe the low-energy ($0 \leq E - E_F << E_F$) excitations of a two-dimensional (2D) system by the standard Hamiltonian (see, for example, Ref. [12])

$$H = H_s + H_d + H^{int}_{d-s} + H^{ext}_{s,d}, \tag{1}$$

in which the expression for the energy of electrons of the s conduction band ("s-electrons") of Ni has the form

$$H_s = H_{kin} + H_{spin}, \quad H_{spin} = \sum_{i,j} J_{dp}(i,j)\, S^z_{i,s} \cdot S^z_{j,s}. \tag{2}$$

The first term in (2) describes the kinetic energy H_{kin} of *s*-electrons, while the second term H_{spin} describes the direct interaction between the spins of two *s*-electrons with the interaction constant

$$J_{dp}(i,j) = \frac{\mu_0 \mu^2}{4\pi} \frac{a^3}{r^3_{ij}}. \tag{3}$$

Here, $S^z_{i,s}$ is the projection of the spin $S_{i,s}$ of the s-electron at the i-th node of a 2D lattice on the z axis; μ_0 is the magnetic permeability; μ is the effective magnetic moment of the spin; a is the lattice constant; and r_{ij} is the distance between electrons at the i-th and j-th nodes of the 2D lattice.

We will not take into account the kinetic energy in the expression

$$H_d = J \sum_{\langle i,j \rangle} S^z_{i,d} \cdot S^z_{j,d} + \sum_{i,j} J_{dp}(i,j)\, S^z_{i,d} \cdot S^z_{j,d} \tag{4}$$

for the energy of electrons in the d-band ("d-electrons"). Because the effective mass of d-electrons is large, we can assume that these electrons are immobile and are localized at the lattice nodes. The first term in (4) describes the interaction between the spins of d-electrons located at the adjacent lattice nodes, while the second term describes the direct interaction between the spins of two d-electrons with the interaction constant $J_{dp}(i,j)$. In (4), J is the constant of the "super-exchange" interaction between adjacent nodes and $S^z_{i,d}$ is the projection of the spin $S_{i,d}$ of the d-electron at the i-th node of the 2D lattice on the z axis.

We will describe the interaction between the s- and d-electrons by the Hamiltonian

$$H_{s-d}^{int} = H_{flip} + \sum_{i,j;b_1 \neq b_2 = s,d} J_{dp}(i,j)\, S_{i,b_1}^{z} \cdot S_{j,b_2}^{z}\,,\ H_{flip} = J_{sd} \sum_{i} S_{i,d}^{z} \cdot S_{i,s}^{z}\,. \tag{5}$$

The first term H_{flip} in (5) describes the exchange interaction between the s- and d-electrons at the same node with the exchange constant J_{sd}, while the second term describes the direct interaction between the spins of the s- and d-electrons localized at the i-th and j-th nodes of the 2D lattice with the interaction constant $J_{dp}(i,j)$. Finally, the term $H_{s,d}^{ext}$ in (1) describes all the rest interactions (Coulomb scattering, interaction with the phonon subsystem and laser field, etc.).

Because $\tau_p >> \tau_{e-ph}, \tau_{e-e}$, we assume that the electron system is thermalized, and will describe the energy distribution of free carriers f_E by the Fermi-Dirac distribution $f_E = \left\{1 + \exp\left[\left(E - E_F\right)/k_B T_e\right]\right\}^{-1}$ [3], where k_B is the Boltzmann constant. We assume that $T_e >> T_{ph}$ during the action of the pumping pulse due to a large thermal conductivity of the lattice (see above), and the spatial distribution of T_e is determined by the intensity distribution I of the radiation. In this case, the solution of the problem is substantially simplified. The movement of s-electrons at distances exceeding the mean free path $l_{free} \cong v_F \tau_{free}$ (v_F is the Fermi velocity and τ_{free} is the mean free path time) can be treated as the diffusion propagation of noninteracting particles in an external self-consistent field specified by H_{spin} and H_{s-d}^{int}. The transition of the electrons from one spin component to another is determined by H_{flip}. The spin flip can be described by the Boltzmann equation, having found the scattering rate in accordance with the Fermi golden rule [3]

$$\left.\partial f_{p,s,\sigma} \middle/ \partial t\right|_{spin-spin}^{out} = -2\pi/\hbar\, f_{p,s,\sigma} \sum_{p',k,k'} \begin{bmatrix} f_{k,d,-\sigma}\left(1 - f_{p',s,-\sigma}\right)\left(1 - f_{k',d,\sigma}\right)\left| M_{p',s,-\sigma;\,k',d,\sigma}^{p,s,\sigma;\,k,d,-\sigma} \right|^2 \cdot \\ \cdot\delta\left(E_{p,s,\sigma} + E_{k,d,-\sigma} - E_{p',s,-\sigma} - E_{k',d,\sigma}\right) \end{bmatrix}. \tag{6}$$

Here, $f_{p,s,\sigma}$ is the distribution function; $M_{p',s,-\sigma;\,k',d,\sigma}^{p,s,\sigma;\,k,d,-\sigma}$ is the matrix element of the spin flip transition between the *s*- and *d*-electrons. Hereafter, we denote the electronic states by triple indices. The first symbol of the index specifies the wave vector (or the energy, see below) of the electron, the second term indicates its belonging to the *s*- or the *d*- band, and the third term describes the orientation of the electron spin. In the random phase approximation [3], which is exact for isotropic bands, the summation over the wave vectors can be reduced to the integration over the energy:

$$\left.\partial f_{p,s,\sigma} \middle/ \partial t\right|_{spin-spin}^{out} = -\tfrac{1}{2} f_{p,s,\sigma} \int_{-\infty}^{\infty} dE'\left(1 - f_{E',s,-\sigma}\right)\rho_s\left(E'\right) W\left(E_{s,\sigma}, E'_{s,-\sigma}\right), \tag{7}$$

$$W\left(E_{s,\sigma}, E'_{s,-\sigma}\right) = \frac{2\pi}{\hbar} \int_{-\infty}^{\infty} d\varepsilon' \rho_d\left(\varepsilon'\right)\rho_s\left(\varepsilon' + \delta E\right) f_{\varepsilon',d,-\sigma}\left(1 - f_{\varepsilon'+\delta E,s,\sigma}\right) \left| M_{(\varepsilon'+\delta E)_{s,-\sigma};E'_{d,\sigma}}^{E_{s,\sigma};\varepsilon_{d,-\sigma}} \right|^2, \tag{8}$$

where $\rho_{s,d}$ is the density of electronic states in the s- and d- bands and $\delta E = E - E'$. For transitions near the Fermi surface, the dependence of the matrix element in (8) on the energy can be neglected, assuming the matrix element equal to $M \cong 0,7$ eV. In this case, the matrix element of scattering is determined by the distribution function for d-electrons and by the density of states. The d-band of transition metals (in particular, Ni) is almost filled, and we can use its parabolic approximation. In this case, we can assume that $\rho_d(E) \propto \sqrt{E_{top} - E}$, where E_{top} is the level of the top of d-band. The density $\rho_s(E) = const$ of the electronic states in the s-band can be assumed constant because the Fermi level is located very far from the bottom of the band.

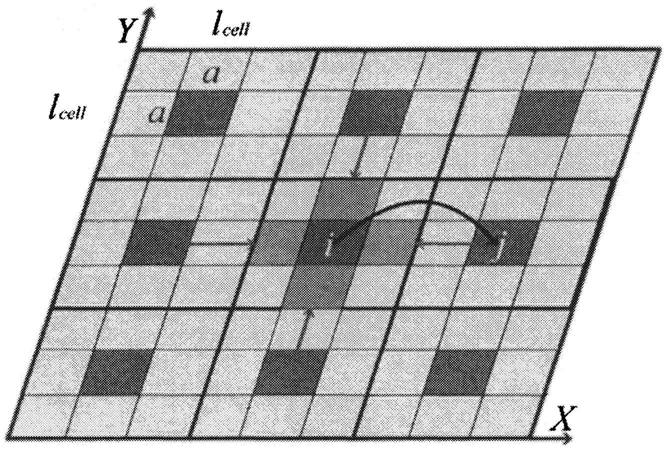

Figure 1. Illustration of the calculation of the interaction of the effective cells in a cluster.

The simulation was performed by the Monte Carlo method [13]. The Ni film was divided by a rectangular grid of 2D clusters with a periodic continuation (Fig. 1). The diffusion constant of s-electrons was assumed to be proportional to the electron velocity $v \cong v_F$, constant, and isotropic. The cell size l_{cell} was chosen equal to the mean free path length l_{free}. In this case, the transition of the s-electron from one cell to another was independent of its previous state, providing the possibility of constructing the Markovian chains. In each cluster, an effective elementary cell was chosen. It was assumed that this cell interacts with effective elementary cells in adjacent clusters as with the next neighbors and that variations in the magnetization at the scale of l_{cell} can be neglected (Fig. 1).

To take into account the direct nonlocal interaction between the spins, which is equivalent to the consideration of the field of the so-called magnetic charges in a classical theory [1], we constructed the Fourier transform of the interaction potential for point sources on the cluster being simulated:

$$G(\mathbf{k}) = \int_{S_{cell}} d\mathbf{r}_{i0}\, J_{dp}(i,0) \exp[\, i\mathbf{k}\mathbf{r}_{i0} \,] \qquad (9)$$

where S_{cell} is the region of integration inside the cluster. Then, taking into account (9), the effective nonlocal interaction potential was written in the form

$$U_{nl}(i) = \int d\mathbf{k} \left[\left(n_{k,s,\sigma} + n_{k,d,\sigma}\right) - \left(n_{k,s,-\sigma} + n_{k,d,-\sigma}\right) \right]^{*} G(\mathbf{k}) \exp[-i\mathbf{k}\mathbf{r}_{i0}\,] \,. \qquad (10)$$

Here, n is the Fourier component of the electron density in the cluster. The diffusion was described by assuming that the *s*-electron moves freely inside the cell, and that the probability of the electron transition into an adjacent cell, which was chosen randomly, is described by the expression [13].

$$P_{ij} = \begin{cases} 1 & \text{for } \Delta E_{ij} \le 0, \\ \dfrac{\exp\left(\Delta E_{ij}/k_B T_e\right)}{\exp\left(\Delta E_{ij}/k_B T_e\right)+1} & \text{for } \Delta E_{ij} > 0. \end{cases} \qquad (11)$$

Here, $\Delta E_{ij} = E_j - E_i$ is the change in the system energy upon the transition from the i-th cell to the j-th cell.

3. RESULTS OF SIMULATION

At the first step, we simulated stationary domains at different temperatures T. The equilibrium domain structure being formed had the maximum ratio of its surface to its perimeter (volume-to-surface ratio). This shows that a positive energy is required for formation of the domain walls. We obtained the temperature dependences for the degree of correlation of the spin orientation for the next neighbors (sites) $C(1) = \left\langle S^z_{0,d} S^z_{1,d} \right\rangle$ (Fig. 2a), the next-next neighbors $C(2) = \left\langle S^z_{0,d} S^z_{2,d} \right\rangle$ (Fig. 2b), and the next-next-next neighbors $C(3) = \left\langle S^z_{0,d} S^z_{3,d} \right\rangle$ (Fig. 2c). One can easily see that the next correlations in the spin orientation are preserved even at $T > T_K$ (Fig. 2a). At the same time, the long-range order disappears at $T = T_K$ (Figs 2b,c). This result completely agrees with calculations performed in paper [14].

The equilibrium domain structure obtained at the first step at room temperature $T_R \cong 300\,K$ was used as the initial condition at the second step of simulation. We considered the influence of the pulse laser heating of the film on its domain structure. We assumed that the film is irradiated by a train of 20-ps pump pulses with the time interval of 5 ns, which considerably exceeds the time of complete cooling of the film down to temperature T_R, while a change ΔT_e in the electron temperature during irradiation is proportional to the incident radiation intensity I. We took into account in the calculations that, unlike the BP technique [9] (spatially uniform heating of the film, $\Delta T_e \cong 500\,K$), in the DFPS [10] the one-dimensional (1D) stationary (at the time scale of τ_p) interference pattern (spatial distribution of the intensity I) is formed in the region of cross-section of two pump beams made coincident in the carrier frequency. We assumed that for the spatial period of this pattern $\Lambda = 5\ \mu$ [10], its visibility factor $\eta = I_{max}/I_{min}$ is ~12, while the maximum change $(\Delta T_e)_{max}$ in the electron temperature at the light-field antinodes is ~900 K. The sequences of the transformation of the simulated domain structure of the film are illustrated in Fig.3 (DFPS) and Fig.4 (BP). The instants of time corresponding to the onset and termination of the N-th ($N = 1 \div 8$) pumping pulse are shown.

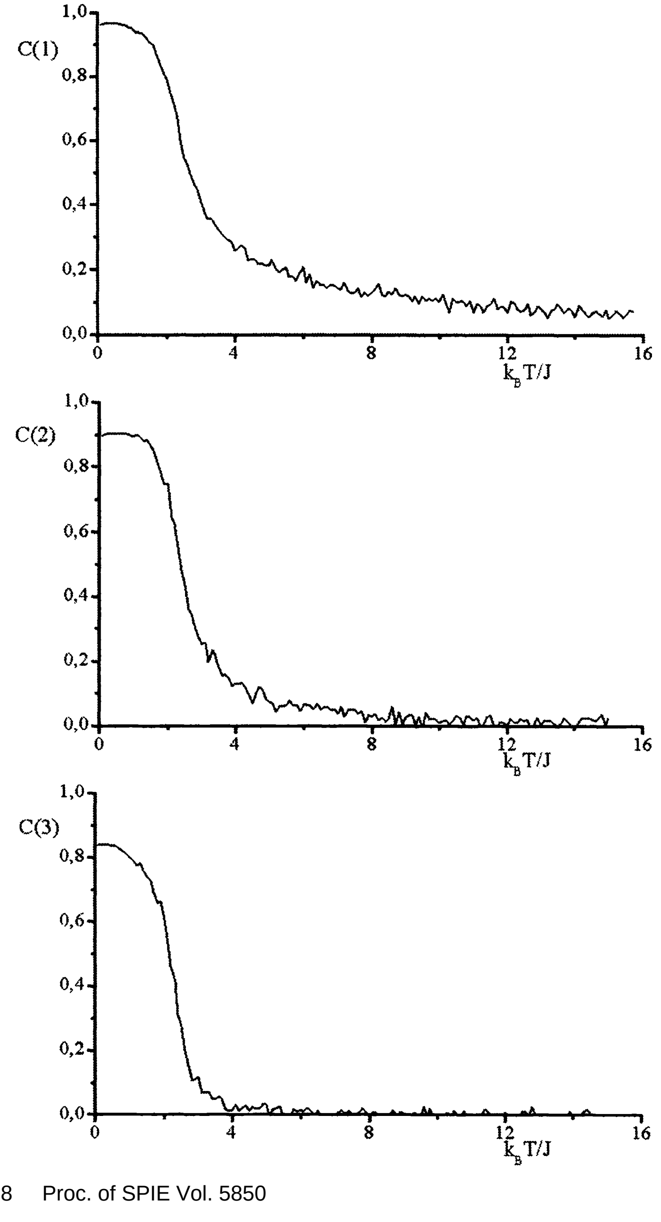

Figure 2. Temperature dependence of the correlation degree in the spin orientation for the next neighbors C(1), the next-next neighbors C(2), and the next-next-next neighbors C(3).

Our simulations showed that after irradiation by only 5-7 pumping pulses, for which $(\Delta T_e)_{max} \cong 900\,K$, the initial domain structure of the film is virtually completely transformed. In DFPS (Fig. 3), the characteristic and very stable periodic (in our calculations, the period was 10 μ) 1D "striped" domain structure has time to form and all stripes are oriented strictly along the interference pattern fringes (Fig. 3). The domain walls very rapidly "adjust" to the dark regions of the interference pattern, and comparatively "cold" ($T_e \ll T_K$ within these regions) free *s*-electrons cannot penetrate through these regions. The period of the domain structure thus formed becomes multiple of Λ. Upon spatially uniform excitation in the BP (Fig. 4), this stabilization factor is completely absent, and the domain structure of the film is almost completely destroyed by the time of termination of the next pumping pulse. Note that, unlike the spatially nonuniform case analyzed here, the transformation of the domain structure of ferromagnetic films caused by spatially uniform optical excitation was already considered in papers [15-18].

The different character of the destruction kinetics of the ferromagnetic film magnetization upon spatially uniform (BP) and nonuniform (DFPS) excitation is quantitatively illustrated by the dependences of C(3) on τ_p plotted by the time of termination of the eighth pump pulse (Fig. 5). One can see from Fig. 5 that the stable domain walls formed upon spatially uniform excitation in the DFPS very efficiently suppress the process of "optical spin mixing" (i.e., the destruction of the domain structure during the spatial migration of excited *s*-electrons between domains). This results in the preservation of the magnetization of the film in DFPS for much longer times than in BP technique.

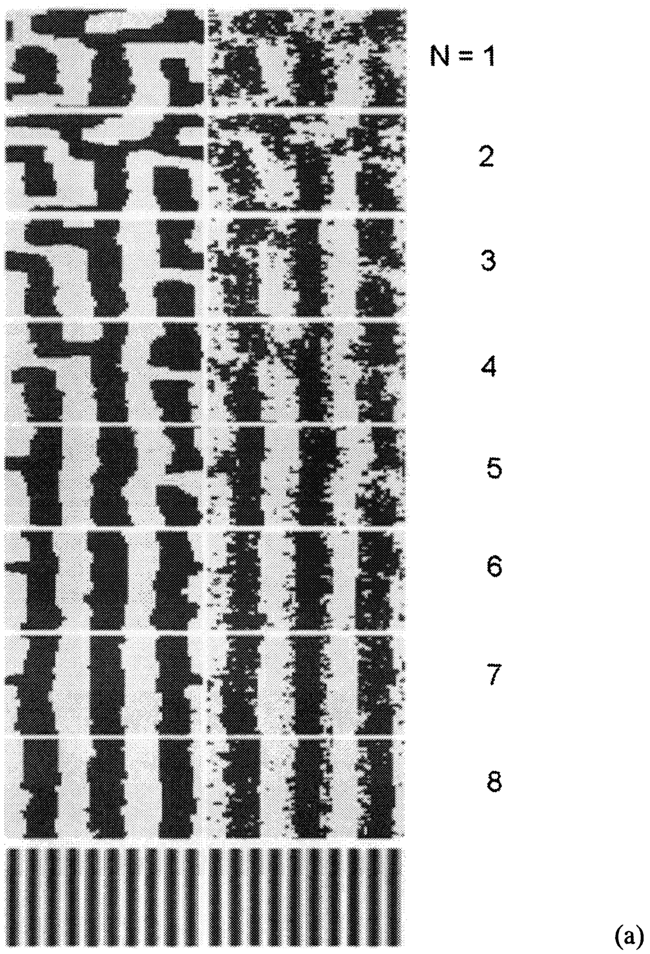

Figure 3. Change in the domain structure of the ultra-thin Ni film in DFPS upon irradiation by a train of 20-ps laser pulses before (left) and after (right) irradiation by the N-th laser pulse. The bright and dark cells have mutually orthogonal orientations of the magnetic moment. The lower figures show localization of bright and dark fringes of the interference pattern.

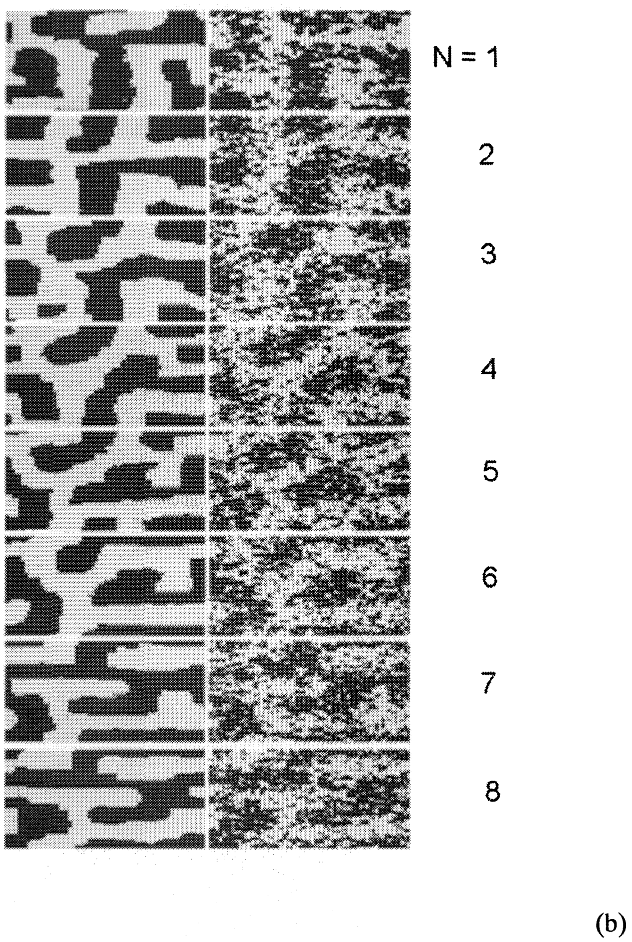

Figure 4. Change in the domain structure of the ultra-thin Ni film in BP upon irradiation by a train of 20-ps laser pulses before (left) and after (right) spatially uniform excitation by the N-th laser pulse. The bright and dark cells have mutually orthogonal orientations of the magnetic moment.

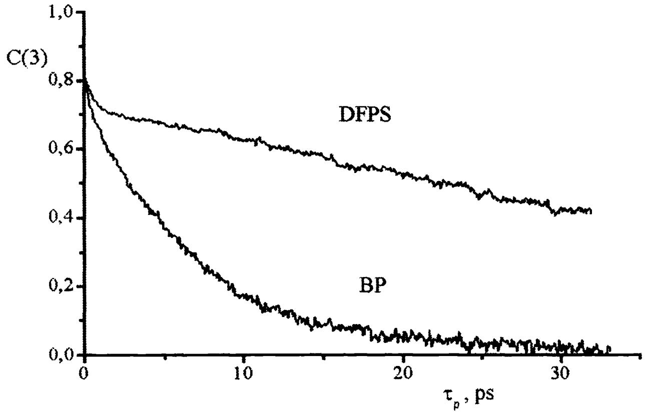

Figure 5. Dependence of the long-range correlation degree C(3) of the spin orientation on the laser irradiation time in DFPS and BP technique.

4. CONCLUSIONS

In the paper, we have shown that remagnetization processes of a ferromagnetic film upon spatially uniform (BP technique [9]) and spatially nonuniform (DFPS [10]) optical excitation are substantially different. Upon spatially nonuniform excitation, the walls of a new stable one-dimensional domain stricture rapidly formed in the film and their positions are rigidly "bound" to the positions of dark fringes of the interference pattern. In this case, spatial migration of excited *s*-electrons is almost completely suppressed. In our opinion, this results in the preservation of the magnetization of ultra-thin Ni films in DFPS [10] during much longer times than in the case of BP technique [9] despite approximately the same intensity of picosecond pumping pulses. However, it is much more important that the considered stabilization gives a possibility to develop a new fast laser technology, based on spatially non-uniform illumination of ultra-thin ferromagnetic films by short trains of ultra-short laser pulses and enabling to create the regular domain structures.

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