

‘Optical spin mixing’ and kinetics of nonlinear response in four-photon spectroscopy of thin ferromagnetic films

A V Voronov, V M Petnikova, K V Rudenko, V V Shuvalov

Abstract. A semiclassical phenomenological model is developed which takes into account basic processes proceeding in experiments on picosecond nonlinear spectroscopy of thin ferromagnetic films. The results of numerical simulations of the evolution of the domain structure of Ni films upon spatially uniform biharmonic pumping (BP) and upon spatially nonuniform excitation in the case of degenerate four-photon spectroscopy (DFPS) are presented. It is shown that the destruction kinetics of the film magnetisation is much slower in the case of DFPS than upon BP. This is explained by the efficient suppression of ‘optical spin mixing’ (the domain-structure destruction due to the spatial migration of s electrons) by potential barriers formed upon spatially nonuniform excitation of stable domain walls.

Keywords: ferromagnetic films, four-photon spectroscopy, spin mixing.

1. Introduction

Interest in processes proceeding in ultrathin ferromagnetic films upon their optical excitation has quickened in the last years due to a rapid development of micro- and nanotechnologies, in particular, due to the necessity of increasing the density and rate of magnetic data recording [1]. To solve these purely applied problems, the ultrafast magnetisation 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 magnetisation kinetics, optical methods are most

efficient. There exist several different opinions in the literature about the demagnetisation rate of ultrathin 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]. This means in fact that magnetisation should relax for the time ~ 100 fs.

Two-photon emission spectroscopy experiments have shown the presence of several demagnetisation processes with characteristic times in the range from 10^{-12} to 10^{-10} s [7]. ‘Direct’ Kerr-effect measurements of the magnetisation [8] have demonstrated that the result of the experiment depends on the pump-pulse duration τ_p . Upon excitation of ferromagnetic films by long laser pulses ($\tau_p > 10$ ns), when the electron temperature T_e should not exceed the Curie temperature T_C , the magnetisation was completely destroyed. However, when 20-ps pump pulses were used, no magnetisation destruction was observed, although in this case the electron temperature T_e should be much higher than T_C .

The results of picosecond ($\tau_p = 20$ ps) nonlinear spectroscopy of ultrathin (~ 17 -nm thick) Ni films by the biharmonic pump (BP) method [9] and of degenerate four-photon spectroscopy (DFPS) [10] also are contradictory. While the DFPS experiments showed that the demagnetisation 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 in fact under the same conditions by two methods of picosecond four-photon spectroscopy, contradict to each other at first glance. We propose a model that takes into account the basic 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 demagnetisation (magnetisation reversal) rates of ferromagnetic films in BP experiments upon spatially uniform excitation and in DFPS experiments upon spatially nonuniform excitation should be substantially different.

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

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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 interest to us, the excited carriers are rapidly thermalised ($\tau_{e-e} \sim 280$ 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 $0 - E_F \ll E_F$, 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 (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 . Based on the experiments [8], whose conditions approximately correspond to the experimental conditions [9, 10], and on paper [11], we will assume in our calculations that the total spin of the system is conserved.

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

$$H = H_s + H_d + H_{s-d}^{\text{int}} + H_{s,d}^{\text{ext}}, \quad (1)$$

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

$$H_s = H_{\text{kin}} + H_{\text{spin}}, \quad H_{\text{spin}} = \sum_{i,j} J_{dp}(i,j) S_{i,s}^z S_{j,s}^z. \quad (2)$$

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

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

Here, $S_{i,s}^z$ is the projection of the spin $S_{i,s}$ of the s electron at the i th site of a two-dimensional 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 sites of the two-dimensional lattice.

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

$$H_d = J \sum_{i,j} S_{i,d}^z S_{j,d}^z + \sum_{i,j} J_{dp}(i,j) S_{i,d}^z S_{j,d}^z \quad (4)$$

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 localised at the lattice sites. The first term in (4) describes the interaction between the spins of d electrons located at the adjacent lattice sites, 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 ‘superexchange’ interaction between adjacent sites and

$S_{i,d}^z$ is the projection of the spin $S_{i,d}$ of the d electron at the i th site of the two-dimensional lattice on the z axis.

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

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

$$H_{\text{flip}} = J_{sd} \sum_i S_{i,d}^z S_{i,s}^z.$$

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

Because $\tau_p \gg \tau_{e-ph}$, τ_{e-e} , we assume that the electron system is thermalised, and will describe the energy distribution of free carriers f_E by the Fermi–Dirac distribution $f_E = \{1 + \exp[(E - E_F)/k_B T_e]\}^{-1}$ [3], where k_B is the Boltzmann constant. We assume that $T_e \gg T_{ph}$ during the action of the pump pulse due to a large thermal conductivity of the lattice (see above), and the spatial distribution of T_e is determined by the distribution of the incident radiation I . In this case, the solution of the problem is substantially simplified. The movement of s electrons at distances exceeding the mean free path $l_{\text{free}} \simeq v_F \tau_{\text{free}}$ (v_F is the Fermi velocity and τ_{free} is the mean free 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]

$$\left. \frac{\partial f_{p,s,\sigma}}{\partial t} \right|_{\text{spin-spin}}^{\text{out}} = -\frac{2\pi}{\hbar} f_{p,s,\sigma} \times \sum_{p',k,k'} \left[f_{k,d,-\sigma} (1 - f_{p',s,-\sigma}) (1 - f_{k',d,\sigma}) \left| M_{p',s,-\sigma;k',d,\sigma}^{p,s,\sigma;k,d,-\sigma} \right|^2 \right. \\ \left. \times \delta(E_{p,s,\sigma} + E_{k,d,-\sigma} - E_{p',s,-\sigma} - E_{k',d,\sigma}) \right]. \quad (6)$$

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:

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

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

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 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 $\rho_d(E) \propto (E_{\text{top}} - E)^{1/2}$, where E_{top} is the top of the d band. The density $\rho_s(E)$ of the electronic states in the s band can be assumed constant because the Fermi level is located far from the bottom of the band.

The simulation was performed by the Monte Carlo method [13]. The entire Ni film was divided by a rectangular net into two-dimensional clusters with a periodic continuation (Fig. 1). The diffusion coefficient of s electrons was assumed to be proportional to the electron velocity $v \simeq v_F$, constant, and isotropic. The cell size l_{cell} was chosen equal to the mean free path 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 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 neighbours and that variations in the magnetisation at the scale of l_{cell} can be neglected (Fig. 1).

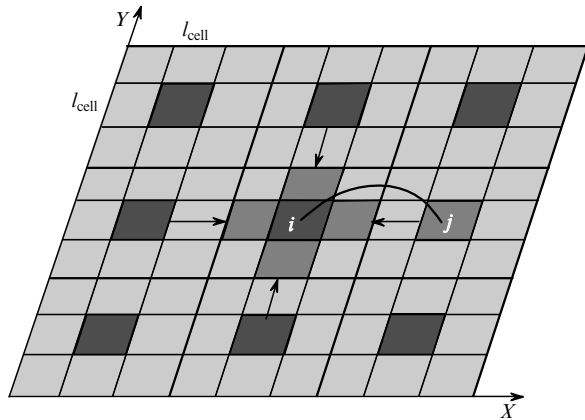


Figure 1. Illustration of the calculation of the interaction of the effective cells in a cluster.

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:

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

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

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

Here, n_{\dots} 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]

$$P_{ij} = \begin{cases} 1 & \text{for } \Delta E_{ij} \leq 0, \\ \frac{\exp(-\Delta E_{ij}/k_B T_e)}{\exp(-\Delta E_{ij}/k_B T_e) + 1} & \text{for } \Delta E_{ij} > 0. \end{cases} \quad (11)$$

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 simulations

At the first stage, 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 neighbours (sites) $C(1) = \langle S_{0,d}^z S_{1,d}^z \rangle$ (Fig. 2a), the next–next neighbours $C(2) = \langle S_{0,d}^z S_{2,d}^z \rangle$ (Fig. 2b), and the next–next–next neighbours $C(3) = \langle S_{0,d}^z S_{3,d}^z \rangle$ (Fig. 2c). One can easily see that the next correlations in the spin orientation are preserved even at $T > T_C$ (Fig. 2a). At the same time, the long-range order disappears at $T = T_C$ (Figs 2b, c). This result completely agrees with calculations performed in paper [14].

The equilibrium domain structure obtained at the first stage at room temperature $T_r \simeq 300$ K was used as the initial condition at the second stage of simulations. 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 method [9] (spatially uniform heating of the film, $\Delta T_e \simeq 500$ K), in the DFPS method [10] the one-dimensional stationary (at the time scale of τ_p) interference pattern (spatial distribution of the intensity I) is formed in the region of intersection of two pump beams made coincident in the carrier frequency. We assumed that for the spatial period of this pattern $\Lambda = 5 \mu\text{m}$ [10], its visibility factor $\eta = I_{\text{max}}/I_{\text{min}}$ is ~ 12 , while the maximum change ΔT_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 (the DFPS method) and Fig. 4 (the BP method). The instants of time corresponding to the onset and termination of the N th ($N = 1 - 8$) pump pulse are shown.

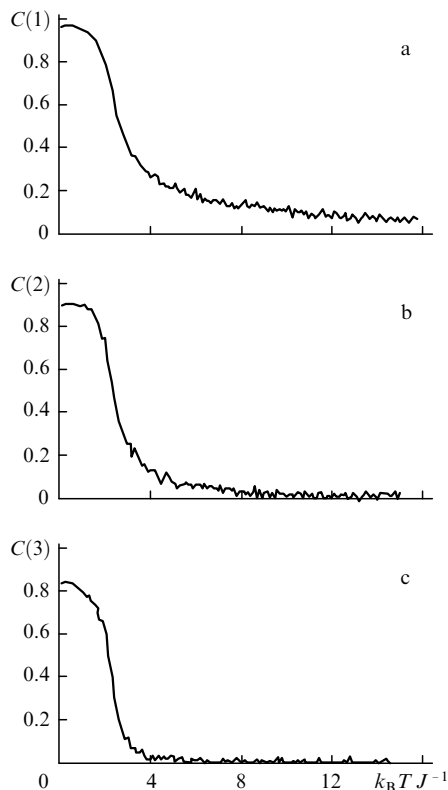


Figure 2. Temperature dependence of the degree of correlation in the spin orientation for the next neighbours $C(1)$ (a), the next–next neighbours $C(2)$ (b), and the next–next–next neighbours $C(3)$ (c).

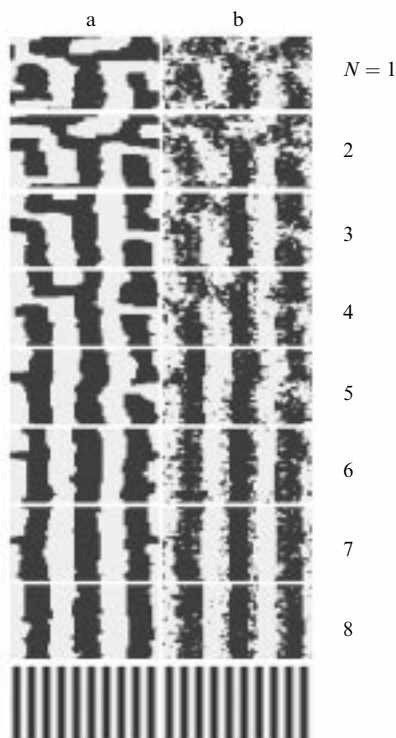


Figure 3. Change in the domain structure of the ultrathin Ni film in the DFPS method upon irradiation by a train of 20-ps laser pulses before (a) and after (b) irradiation by the N th laser pulse. The bright and dark cells have mutually orthogonal orientations of the magnetic moment. The lower figures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

Our simulations showed that after irradiation by 5–7 pump pulses, for which $\Delta T_e^{\max} \simeq 900$ K, the initial domain structure of the film is virtually completely transformed. In the DFPS method (Fig. 3), the characteristic and very stable

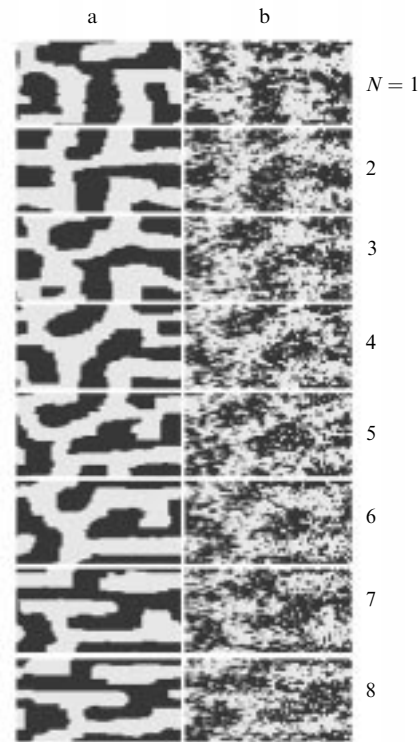


Figure 4. Change in the domain structure of the ultrathin Ni film in the BP method upon irradiation by a train of 20-ps laser pulses before (a) and after (b) spatially uniform excitation by the N th laser pulse. The bright and dark cells have mutually orthogonal orientations of the magnetic moment.

periodic (in our calculations, the period was $10 \mu\text{m}$) one-dimensional ‘band’ domain structure has time to form, which is oriented strictly along the interference pattern fringes (Fig. 3). The domain walls rapidly ‘adjust’ to the dark regions of the interference pattern, and comparatively ‘cold’ ($T_e \ll T_C$ 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 method (Fig. 4), this stabilisation factor is completely absent, and the domain structure of the film is almost completely destroyed by the time of termination of the next pump pulse. Note that, unlike the spatially nonuniform case analysed 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 magnetised ferromagnetic film upon spatially uniform (the BP method) and nonuniform (the DFPS method) 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 method 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 do-

mains). This results in the preservation of the magnetisation of the film in the DFPS method for much longer times than in the BP method.

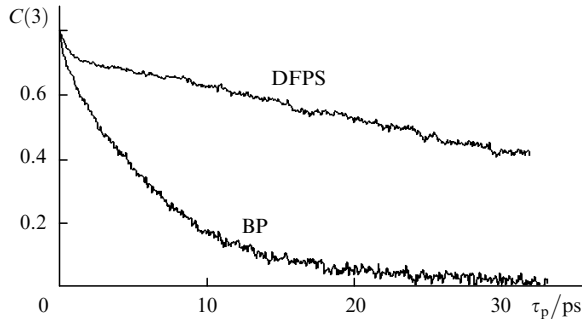


Figure 5. Dependence of the degree of long-range correlations $C(3)$ of the spin orientation on the laser irradiation time in the DFPS and BP methods.

4. Conclusions

We have shown in this paper that the destruction kinetics of the magnetisation of a ferromagnetic film upon spatially uniform (the BP method [9]) and spatially nonuniform (the DFPS method [10]) optical excitation are substantially different from each other. Upon spatially nonuniform excitation, the walls of a new stable one-dimensional domain structure rapidly formed in the film, whose positions are rigidly bound to the positions of the dark fringes of the interference pattern, almost completely suppress the spatial migration of excited s electrons. In our opinion, this results in the preservation of the magnetisation of ultrathin Ni films in the DFPS method [10] during much longer times than in the case of the BP method despite approximately the same intensity of picosecond pump pulses.

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