# The indispensable role of the transversal spin fluctuations mechanism in laser induced demagnetization of Co/Pt multilayers with nanoscale magnetic domains

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**Abstract**

The switching of magnetic domains induced by ultrashort laser pulse has been demonstrated in nanostructured ferromagnetic films. It leads to the dawn of a new era for breaking the ultimate physical limit for the speed of magnetic switching and manipulation, which is relevant to current and future information storage. However, the understanding for the interactions between light and spins is still lacking in magnetic heterostructures with nanoscale domain structures. Here, both the time resolved magneto-optical Kerr (TRMOKE) experiments and atomistic simulations were carried out to investigate the dominant mechanism of laser-induced ultrafast demagnetization in $[Co/Pt]_{20}$ multilayers with nanoscale magnetic domains. It is found that the ultrafast demagnetization time keeps as a constant value with various magnetic configurations, indicating that the domain structures play a minor role in laser induced ultrafast demagnetization. In addition, both in experiment and atomistic simulations, we find a dependence of the behavior of ultrafast demagnetization time $\tau_M$ on the laser fluence, which is in contrast to the observations of spin transport within magnetic domains.The remarkable agreement between experiment and atomistic simulations indicates that the local dissipation of spin angular momentum is the dominant demagnetization mechanism in this system. More interestingly, we made a comparison between atomistic spin dynamic simulation and the longitudinal spin flip model, highlighting that the transversal spin fluctuations mechanism is responsible for the ultrafast demagnetization in the case of inhomogeneous magnetic structures. This is a significant progress in clarifying the microscopic mechanism

underlying the process of ultrafast demagnetization in the inhomogeneous magnetic structures.

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## 1. Introduction

The ferromagnetic thin films with nanoscale domain structures have attracted considerable attention due to its potential to serve as the low-power spintronic devices[1,2]. In the past decades, the magnetic field-driven domain wall motion[3] in Co/Pt multilayers with strong perpendicular magnetic anisotropy has been extensively reported as well as the current-induced domain wall motion via the spin transfer torque[4]. Other techniques including the electric field[5], voltage-induced strain[6] and thermal gradient[7] have also been utilized to manipulate the nanoscale magnetic domain structures. The discovery of ultrafast demagnetization, first reported by Beaurepaire et al[8]. in 1996, opened up new routes for manipulating magnetization on the sub-picosecond timescale. For instance, an important milestone from the studies of the ultrafast spin dynamics is the observation that the ultrashort laser can directly switch the magnetic domains in ferrimagnetic GdFeCo[9] without an external field. It leads to the dawn of a new era for breaking the ultimate physical limit for the speed of magnetic switching and manipulation. Recently, such all optical switching has extended to the ferromagnetic Co/Pt multilayers as well as the FePt nanoparticles[10]. However, apart from the demonstrated potential technologies for heat-assisted magnetic recording (HAMR), the investigations of the fundamental interactions between spins, electrons and lattices far from equilibrium are still lacking in the case of Co/Pt heterostructures with nanoscale magnetic domain configurations.

Since 1996, significant progress in understanding the microscopic mechanism of ultrafast spin dynamics including the important role of spin-orbit coupling[11], the direct

interaction between spins and photons[12] as well as the spin transport[13]in the multilayer thin films has been achieved so far. In hindsight, most of these reports have focused on magnetic media with single domain structures[14]. In the case of inhomogeneous magnetic domain structures, the spin transport between neighboring magnetic domains has been demonstrated with the advent of femtosecond-pulse X-ray sources[15,16] in $[Co/Pd]_{30}$ multilayer films as well as in $[Co/Pt]_{16}$ structures. However, Moisan et al cannot exclude the contributions from local spin flip scattering[17] by means of time resolved magneto-optical Kerr effect (TRMOKE). In fact, the local approach[18, 19] such as the plain three-temperature model (3TM)[8] qualitatively describes the intense laser induced temperature evolution of the electrons, lattice, and spins with time. Based on this model, atomistic Landau-Lifshitz-Gilbert (LLG) method[20, 21, 22] with Langevin dynamics is capable of reproducing the rapid decrease of the magnetization observed in experiment. In this case, the ultrashort laser pulse excitation leads to a non-equilibrium divergence between the electron temperature $T_e$ and lattice temperature, $T_l$. We treat the electron gas as the heat bath for the spin system. Moreover, the conserved spin angular momentum is transferred locally and represented by the phenomenological Gilbert damping parameter.[23,24] This computational model ignores the specific angular momentum transfer channel, whilst it provides a straightforward way[25] to understand the physics underlying the temporal evolution of magnetization after laser pulse excitation.

Considering that a consensus as to the dominant mechanism responsible for ultrafast demagnetization is still lacking in the multilayers with nanoscale magnetic

domains, in this paper, the magnetic domain configuration dependent ultrafast demagnetization curves have been obtained via time-resolved magneto-optical Kerr effect (TRMOKE) experiment in Co/Pt multilayers. Both in experiment and atomistic spin dynamics simulations, the laser fluence dependent ultrafast demagnetization curves have been produced to demonstrate the indispensable role of local spin angular momentum dissipation in the presence of magnetic domain configurations. The evolution of ultrafast demagnetization time $\tau_M$ as functions of Gilbert damping has been compared between atomistic spin dynamics simulation and longitudinal spin flip model. Based on this comparison, the explicit mechanism of local spin angular momentum dissipation in the case of inhomogeneous magnetic structures is illustrated clearly in the simulation model, which is a significant progress in understanding the ultrafast demagnetization mechanism in Co/Pt system with magnetic domains structures.

## 2. Experimental

### 2.1 Experimental method

In this study, both the applied field and laser fluence dependent ultrafast demagnetization curves for Ta(5 nm)/Pt(2 nm)/[Co(0.4 nm)/Pt(0.7 nm)]$_{20}$/Pt(2.3nm) multilayers have been achieved by using time-resolved magneto-optical Kerr effect (TRMOKE) technique[14,26]. A train of optical pulses with a wavelength of 780 nm, 55 fs duration and 100 nJ/pulse is generated at 5.2 MHz repetition rate by a Ti: sapphire oscillator (FEMTOLASER, XL-100). A 200μm thickness BBO crystal was used to

double the frequency of femtosecond laser. The laser beam from the source is split into both 780 nm and 390 nm beams. We use the 780 nm laser as the pump pulse to excite the magnetic system out of equilibrium, while the 390 nm laser pulse was used as a probe beam to measure the subsequent magnetization dynamics with the timescale from sub-picosecond to nanosecond. The pump laser beam is much stronger than the probe with an intensity ratio of at least 20 for the lowest pump fluence. Both the pump and probe beam are incident along the normal axis (*z*-axis) of the sample. The detection geometry is only sensitive to the out-of-plane component of the magnetization $M_z$. The pump and probe beams are focused onto the sample with spot diameters of ~10 μm and ~5 μm via a $20\times$ objective lens, respectively.

**2.2 The measurements of static properties and spin precession for Ta (5 nm)/Pt (2 nm)/[Co (0.4 nm)/Pt (0.7 nm)]$_{20}$/Pt (2.3 nm).**

The sample used in this study is a 22 nm [Co(0.4 nm)/Pt(0.7 nm)]$_{20}$ multilayer thin film, grown at room temperature by dc magnetron sputtering[27]. As shown in Fig. 1, the hysteresis loop along the surface normal of the film is measured by Vibrating Sample Magnetometer (VSM). It is found that the Co/Pt multilayer exhibits an out-of-plane magnetic anisotropy, and an obvious jump in the loop occurs even before the applied field is reversed. The jump mainly comes from the onset of the domain formation, which is illustrated in Fig. 1(b) by the measurement of Lorentz TEM showing the 260 nm domain structure.

To obtain the effective magnetic anisotropy, we performed the laser-induced magnetization precession experiment. In this case, the external field *H* ranged from

2.5 kOe to 4.3 kOe was applied at $\theta_H$=80° from the normal direction of the sample. The typical time-resolved magnetization dynamics with various applied fields shown in Fig. 2(a) can be fitted by the damped harmonic function added to an exponential decaying background[28]:

$$\Delta M(t) = A + B\exp(-\nu t) + C\exp(-\frac{t}{\tau})\sin(2\pi ft + \varphi) \quad (1)$$

Where $A$ and $B$ are the background magnitudes, and $\nu$ is the background recovery rate. $c, \tau, f$ and $\varphi$ are the magnetization precession amplitude, relaxation time, frequency and phase, respectively. From the fitting curves shown in Fig.2 (a) as the solid lines, the value of precession frequency $f$ is extracted. Fig. 2 (b) shows the frequency as a function of applied field. The experimental $f - H$ relation can be fitted by analytic Kittel formula derived from LLG equation:

$$f = \frac{\gamma}{2\pi}\sqrt{H_1 H_2} \quad (2)$$

Where $H_1 = H\cos(\theta_H - \theta) + H_K^{eff}\cos^2\theta$, $H_2 = H\cos(\theta_H - \theta) + H_K^{eff}\cos 2\theta$

The equilibrium angle of magnetization was obtained from the relationship $\sin 2\theta = \frac{2H}{H_K^{eff}}\sin(\theta_H - \theta)$. And the direction of applied field is fixed at $\theta_H = 80^\circ$. In the above equations, $H_K^{eff}$ and $\gamma$ are the effective perpendicular magnetization anisotropy and gyromagnetic ratio, respectively, where $H_K^{eff} = \frac{2K_{eff}}{M_s}, \gamma = \frac{2\pi g\mu_B}{h}$. In our calculation, the Lande $g$-factor was set to 2.2 as the bulk Co value, and the best fitting value of $K_{eff}$ is $2.8\times10^6 erg/cm^3$ for [Co/Pt]$_{20}$ multilayer[29]. We take this value as the input parameter in the atomistic simulation below.

**2.3 The measurements of ultrafast demagnetizaion curves for Ta (5 nm)/Pt (2 nm)/[Co (0.4 nm)/Pt (0.7 nm)]$_{20}$/Pt (2.3 nm).**

In previous studies[15, 16], the femtosecond-pulse X-ray sources have been used to demonstrate the acceleration effect of spin angular momentum transferring between neighboring domains on ultrafast demagnetization. This makes the role of magnetic domain structures played in ultrafast demagnetization interesting. In order to clarify the role of spin transport played in various domain configurations, we carried out the time-resolved MOKE measurements for different applied fields.When $H$ is above 400 Oe, the sample is completely magnetized. With reducing the applied fields from a saturated one of 900 Oe, the multi-domain configurations appear gradually. Fig. 3(a) shows the magnetization as a function of time delay for a series of magnetic domain configurations at a fixed incident laser fluence of 0.5 mJ/cm$^2$. We can clearly observe that the evolution of magnetization curves looks identical for various applied fields. The solid lines reproduce the experimental data by the three temperature model (3TM model) as follows[30]:

$$-\frac{\Delta M(t)}{M} = \left\{\left[\frac{A_1}{\left(t/\tau_0+1\right)^{0.5}} - \frac{\left(A_2\tau_E - A_1\tau_M\right)}{\tau_E-\tau_M}e^{-\frac{t}{\tau_M}} - \frac{\tau_E(A_1-A_2)}{\tau_E-\tau_M}e^{-\frac{t}{\tau_M}}\right]\Theta(t) + A_3\delta(t)\right\} * G(t,\tau_G) \quad (3)$$

$G(t,\tau_G)$ presents the Gaussian laser pulse profile, whose full width at half maximum (FWHM) is $\tau_G$. $\Theta(t)$ is a step function, $\delta(t)$ is the Dirac delta function. $\tau_M$ is defined as the time needed for magnetization to reach a level of $(1-e^{-1})$ of its maximum demagnetization[24] and $\tau_E$ is the electron-phonon equilibration time,

describing the rate at which electrons and phonons exchange their energy and reach a temperature equilibrium[31]. The parameter $\tau_0$ represents the heat transport timescale through the substrates. In this model, the electrons absorb the laser photons directly, and then create the hot electrons. Once the thermalization is produced by Coulomb interactions, the electrons, spins and phonons can be described by its own temperature. The relaxation takes place through energy transfer between different baths. Although it ignores the angular momentum transferring, the 3TM model has been widely used to extract the ultrafast demagnetization time. Eq.(3) is solved based on a set of differential equations (4) of 3TM model by neglecting the spin specific heat in the low fluence limit[32].

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$$\begin{aligned} C_e \frac{\partial T_e}{\partial t} &= -G_{e-p}(T_e - T_p) - G_{e-s}(T_e - T_s) + S(t) \\ C_p \frac{\partial T_p}{\partial t} &= -G_{e-p}(T_p - T_e) - G_{s-p}(T_p - T_s), \\ C_s \frac{\partial T_s}{\partial t} &= -G_{e-s}(T_s - T_e) - G_{s-p}(T_s - T_p) \end{aligned} \tag{4}$$

where C are the heat capacities of the three systems, G the coupling constants between spin, electron and phonon, and S(t) represents the excitation from the laser pump pulse. Fig. 3(b) shows that the demagnetization time $\tau_M$ is a constant value with various applied fields, indicating that there is no obvious influence of the domain structures on ultrafast demagnetization time. It is consistent with what has been observed by TRMOKE experiment in both Co/Pd and Co/Pt multilayers with magnetic domains[17].

In the case of magnetic domain structures, the spin transport induced ultrafast

demagnetization time $\tau_M$ is independent of the laser fluence[15]. It is completely different from the previous results based on local spin-flip scattering[33, 34]. Therefore, we performed the time resolved measurements as a function of laser fluence at H = 50 Oe, as shown in Fig. 4 (a). We chose such a value of applied field because it leads to a multidomain state as demonstrated in Fig. 1. It is obvious that the higher laser fluence gives rise to a longer time needed to demagnetize the sample as the maximum magnetic quenching increases. Because the Eq.(3) is valid in the low fluence limit, the largest laser fluence used here is 2 mJ/cm$^2$. The demagnetization curves obtained in a laser fluence larger than 2 mJ/cm$^2$ would not be reproduced well by Eq.(3) giving rise to the invalid value of demagnetization time $\tau_M$. We have to address that the critical value of the laser fluence differs largely within different systems. It is mainly due to various thermal conductivity of the samples[17]. Fig. 4 (b) reports the demagnetization time $\tau_M$ extracted from the 3TM model with various laser fluence as well as $\tau_E$ charactering the magnetization recovery time. An almost linear relation between the demagnetization time $\tau_M$ and laser fluence is established. Moreover, the values of $\tau_M$ fall into the range of 150~300 fs which agrees very well with that obtained in a 15 nm thick homogeneous Co film, where it was explained by Koopmans et al. using electron-phonon medicated spin-flip scattering model[35]. The close laser fluence dependence of the demagnetization time provides further evidence that the spin-flip scattering dominates the ultrafast demagnetization in the present system. In addition, the recovery time $\tau_E$ of magnetization slows down obviously by increasing the laser fluence, agreeing with previous results[36] obtained by both experiment as well as

microscopic LLB calculations.

Our conclusions contrast with the previous demonstration in Co/Pt multilayers using femtosecond-pulse X-ray sources[15], in which the hot electrons displacement between neighboring domains plays a major role in the ultrafast demagnetization process. The different lifetimes as well as velocities between spin-majority and spin-minority hot electrons can induce an imbalance spin accumulation in the region close to the domain wall, resulting in the local magnetization quenching. The estimated spin transport induced domain wall broadening is around 20 nm[17]. Therefore, in the future, reducing both the spatial resolution of laser source and the domain size in the samples can facilitate the exploration of spin dependent hot-electron transport in ferromagnets with nanoscale magnetic domains. However, the explicit mechanism of local spin angular momentum dissipation in such inhomogeneous system with nanoscale magnetic domains has never been mentioned so far. This is precisely the central strategy in this paper.

## 3. Atomistic spin dynamics model

### 3.1 Simulation method

The atomistic spin dynamics simulations[20, 21]was performed using the VAMPIRE software to investigate the microscopic mechanism underlying ultrafast demagnetization. In this atomistic simulation, the spin Hamiltonian $\Im$ of the systems are described by an extended Heisenberg spin model with the following form:

$$\Im = -\sum_{i \neq j} J_{ij} \overrightarrow{S_i} \bullet \overrightarrow{S_j} - K_{eff} \sum_i (\overrightarrow{S_i} \bullet \vec{e})^2 - \sum_i \mu_s \overrightarrow{S_i} \bullet \overrightarrow{H_{app}} \qquad (5)$$

The first term is the Heisenberg exchange energy, where $J_{ij} = 6.064 \times 10^{-21} J / link$

is the exchange interaction constant between the nearest neighboring two spins $\overrightarrow{S_i}$ and $\overrightarrow{S_j}$. The second term describes the magnetocrystalline anisotropy of the spin, where $K_{eff} = 1.1 \times 10^{-24} J/atom$ is the effective uniaxial anisotropy constant induced mainly by Co/Pt interface. The last term is the Zeeman energy involving interactions between the system and external applied fields, where $\mu_s = 1.72\mu_B$ is the magnetization moment per atom. The dynamics of spin systems are determined by the Landau-Lifshitz-Gilbert equation with Langevin dynamics:

$$\frac{\partial \overrightarrow{S_i}}{\partial t} = -\frac{\gamma}{(1+\lambda^2)}[\overrightarrow{S_i} \times \overrightarrow{H^i{}_{eff}} + \lambda \overrightarrow{S_i} \times (\overrightarrow{S_i} \times \overrightarrow{H^i{}_{eff}})] \tag{6}$$

where $\gamma$ is the gyromagnetic ratio, $\lambda$ is the microscopic Gilbert damping parameter mainly coming from intrinsic contributions of spin-electron and spin-lattice interactions. $\overrightarrow{H^i{}_{eff}}$ is the net magnetic field on each spin including an additional white noise term:

$$\overrightarrow{H^i_{th}} = \Gamma(t)\sqrt{\frac{2\lambda k_B T}{\gamma \mu_s \Delta t}},$$

where $k_B$ is the Boltzmann constant, $T$ is the temperature of electron system, $\Delta t$ is the integration time step, and $\overrightarrow{\Gamma(t)}$ is the Gaussian white noise term representing the thermal fluctuations on each atomic site. So, the effective field in the LLG equation with Langevin Dynamics reads:

$$\overrightarrow{H^i_{eff}} = -\frac{1}{\mu_s}\frac{\partial \Im}{\partial \overrightarrow{S_i}} + \overrightarrow{H^i_{th}}$$

The electron system temperature is calculated from a two-temperature model[20]:

$$C_e \frac{\partial T_e}{\partial t} = -G_{e-p}(T_e - T_p) + S(t)$$
$$C_p \frac{\partial T_p}{\partial t} = -G_{e-p}(T_p - T_e) \quad (7)$$

Where $C_e, C_p$ are the electron and lattice heat capacities, respectively. $T_e$ is the electron temperature, $T_p$ is the lattice (phonon) temperature, $G_{e-p}$ is the electron-lattice coupling constant, and the parameter $S(t)$ is determined by a Gaussian pulse with height proportional to the effective laser fluence via the relationship as follows:

$$S(t) = F_{eff} \bullet e^{-\frac{(t-3t_0)^2}{t_0}}, \quad (8)$$

where $F_{eff}$ is the effective laser fluence parameter with non-dimension and $t_0$ is the duration of the laser pulse. The time evolution of the electron temperature is solved using a simple Euler scheme.

In the numerical simulation carried out by Vampire, we assume that the heat capacity of lattice $C_p$ is independent of the lattice temperature and given by $C_p = 8.5 \times 10^6 J \bullet m^{-3} \bullet K^{-1}$, while the electronic heat capacity $C_e$ is taken proportional to the temperature $T_e$ via $C_e = \gamma T_e$ with $\gamma = 3 \times 10^3 J \bullet m^{-3} \bullet K^{-1}$. The value of electron-lattice coupling parameter $G_{e-p}$ is set as $1.5 \times 10^{18} W \bullet m^{-3} \bullet K^{-1}$. The values of all the parameters are consistent with those in literatures[8, 23, 32]. In addition, the value of effective laser fluence $F_{eff}$ was increased from $6 \times 10^{20}$ to $5 \times 10^{21}$ monotonously in the numerical calculations to reproduce the experimental curves with laser fluence increasing from 0.5 mJ/cm$^2$ to 2 mJ/cm$^2$ in Fig.4(a).

**3.2 Simulation results and discussions**

To demonstrate the indispensable role of the local spin angular momentum

dissipation in this system, the atomistic simulations were carried out to reproduce the laser fluence dependent experimental curves. As shown in Fig. 4 (c), we can clearly observe that the experimental curves are reproduced exactly within the atomistic spin model by increasing the laser power, justifying the local spin angular momentum dissipation suffices here to explain the ultrafast demagnetization. In the case of F = 2 mJ/cm$^2$, the simulation result shown as the dashed red line disagrees with the experimental curve. This discrepancy can be attributed to the nonlinear temperature dependence of electronic heat capacity in reality, which always takes place at high laser fluence[37]. However, this effect is ignored in the current simulation model. In this case, a larger value of $\gamma = 6.2 \times 10^3 J \bullet m^{-3} \bullet K^{-2}$ could be used in the simulation to reproduce the experimental curve as is shown by the solid red line in Fig. 4(c). On the other hand, the effect of heat accumulation is more pronounced with the laser fluence increasing. It can be demonstrated as the recovery time $\tau_E$ increases with the laser fluence increasing in Fig. 4(b). However, such effect is also not considered in the simulation model, which may be another reason for the deviation of the simulated result from the experimental one. Despite this, the atomistic calculations reproduce the main features in TRMOKE experiment, namely, an increase of the demagnetization time is needed when the loss of magnetization increases. As shown in Fig. 4(d), the increasing laser fluence results in an increase of the electron temperature. The higher electron temperature leads to a larger maximal demagnetization. Consequently, a longer relaxation time is needed to demagnetize the system.

In the present atomistic spin model, the microscopic damping parameter[20] $\lambda$ which represents the local intrinsic contributions from spin-lattice and spin electrons interactions is used to account for the local spin angular momentum transfer to induce ultrafast demagnetization. To highlight the microscopic mechanism responsible for ultrafast demagnetization in the present system, we address the relationship between ultrafast demagnetization time and microscopic Gilbert damping parameter, since both of them require a transfer of angular momentum from the electronic system to the lattice. In the case of 3*d* transition metal Cobalt shown as the dotted line in Fig. 5(a), at a given laser fluence, we can clearly note that the maximum magnetic quenching increases as the microscopic Gilbert damping parameter increases, while the demagnetization time reduces. In fact, the microscopic Gilbert damping parameter $\lambda$ coming from the local intrinsic contributions (spin-lattice and spin-electron interactions) in the atomistic spin dynamics model, as the bridge between the spins and the heat baths of electrons and phonons, represents the strength of spin-orbit coupling effect[38]. Therefore, it is expected that a larger microscopic Gilbert damping $\lambda$ can make the demagnetization faster and larger as shown in Fig. 5(a). This agrees qualitatively with the prediction given by phonon-mediated spin-flip scattering, where the spin orbit coupling effect induces the spin mixing probability[39] and consequently the spin flip scattering.

Despite this agreement, to illustrate the mechanism of ultrafast demagnetization in the framework of atomistic simulations, Fig. 5(b) highlights the difference of ultrafast demagnetization time $\tau_M$ between the atomistic simulations and the

longitudinal spin flip model by Koopmans et al[19]., as functions of the microscopic Gilbert damping $\lambda$. In the case of longitudinal spin flip model, an inverse relation shown as the red line in Fig. 5(b) between $\tau_M$ and $\lambda$ has been derived via the Curie temperature $T_c$, $\tau_M = C_0 \frac{h}{2\pi K_B T_c \lambda}$, with $h$ and $K_B$ the Plank and Boltzmann constants, respectively. $C_0 = \frac{1}{4}$ is a constant value determined by Elliot-Yafet type scattering[19]. The atomistic simulation results are fitted by the 3TM model shown as the solid lines in Fig. 5(a), from which we extract the value of the demagnetization time. The extracted demagnetization time $\tau_M$ as a function of $\lambda$ is shown in Fig. 5(b) as the black dots. An exponential function is used to reproduce the relationship between $\tau_M$ and $\lambda$ obtained from the atomistic simulations and indicates a more gradual change of the ultrafast demagnetization time with Gilbert damping compared with that given by longitudinal spin-flip scattering（phonon-mediated Elliot-Yafet type） shown as the red line in Fig. 5(b). The difference mainly comes from the fact that the transverse spin fluctuations determine the ultrafast demagnetization in the atomistic spin dynamics simulations, where the length of the local spin moment is fixed. This is contrast to the model used by Koopmans *et al*., in which the magnitude of atomic moment is reduced by longitudinal spin-flips in Elliot-Yafet scattering events[35]. In fact, transverse spin fluctuations[22] have been demonstrated as the possible explanation of ultrafast demagnetization by TRMOKE experiment[40] as well as spin resolved two-photo photoemission techniques[41].

Indeed, the Ref. 34 also reported the similar inverse relation between $\lambda$ and $\tau_M$ within the micromagnetic Landau-Lifshitz-Bloch (LLB) model. The LLB

equation treats both transverse and longitudinal fluctuations of the atomic magnetic moments. It contains two parameters, a transverse and a longitudinal relaxation parameter which both are related to the intrinsic coupling-to-the bath parameter $\lambda$. This coupling parameter can be related to the actual matrix elements for spin-flip scattering. In contrast, only the transverse relaxation is involved in atomistic LLG model used in this study. Despite this, the consistent results obtained in both LLB and LLG equations indicate that the phenomenological equations applied both at micromagnetic and atomistic scales contain the physics of ultrafast demagnetization behavior. Due to the lack of contributions from longitudinal relaxation to ultrafast demagnetization in atomistic spin model, the comparison was made between atomistic spin model and longitudinal spin flip model by establishing the explicit relationship between Gilbert damping constant and ultrafast demagnetization time. Thereby, we proposed that the transversal spin fluctuations is responsible for the ultrafast demagnetization mechanism in the current system.

**4. Conclusions**

In this study, laser induced ultrafast demagnetization dynamics in $[Co/Pt]_{20}$ multilayers with magnetic domain configurations has been studied using both TRMOKE experiment and atomistic spin dynamics simulations. It is found experimentally that the demagnetization time $\tau_M$ keeps a constant value of 150 fs with various magnetic domain structures, justifying that the spin dependent hot electron transport between neighboring domains plays a minor role in ultrafast

demagnetization in our samples. Moreover, the experimental evidence for a local spin-flip scattering mechanism, namely, the demagnetization time increases with the laser fluence increasing, is reproduced exactly by an atomistic spin dynamics simulation based on the model of local spin angular momentum dissipation. Via atomistic spin dynamics model, the transversal spin fluctuations mechanism has been demonstrated to be responsible for the ultrafast demagnetization in the case of Co/Pt multilayers with inhomogeneous magnetic structures. This is a significant progress in clarifying the microscopic mechanism underlying the ultrafast demagnetization in the inhomogeneous magnetic structures.

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**Figure caption:**

**Figure 1. (color online) Static magnetic properties of Ta(5 nm)/Pt(2 nm)/[Co(0.4 nm)/Pt(0.7 nm)]$_{20}$/Pt(2.3nm) multilayers. (a)** The hysteresis loop along the perpendicular direction of the sample measured by VSM with the maximum applied fields of 4 kOe. (b) Lorentz TEM images measured at zero applied field. The side images are the zoom-in for the domain structure in dashed yellow box. Un, in and ov represent under-focused, in-focused and over-focused L-TEM images separately.

**Figure 2. (color online). (a) TRMOKE signals for Ta(5 nm)/Pt(2 nm)/[Co(0.4 nm)/Pt(0.7 nm)]$_{20}$/Pt(2.3nm) multilayers with applied fields H = 2500 Oe, 2900 Oe, 3700 Oe, 3900 Oe, 4300 Oe. (b) Magnetic field dependence of precession frequency with magnetic field applied at $\theta_H$=80° from the normal direction of the sample.**

**Figure 3. (color online). Ultrafast demagnetization curves and demagnetization time.** (a) Ultrafast demagnetization curves as a function of applied fields with H = 0 Oe, 50 Oe, 100 Oe, 300 Oe, 600 Oe, 900 Oe. (b) Extracted demagnetization time as a function of applied fields.

**Figure 4. (color online). TRMOKE experimental and atomistic simulation results.** (a) Ultrafast demagnetization curves with various laser fluences ranging from 0.5 mJ/cm$^2$ to 2 mJ/cm$^2$. The solid lines represent the fitting data by 3TM. (b) Extracted demagnetization time from 3TM as well as recovery time as a function of laser fluences. (c) The experimental demagnetization curves reproduced by atomistic simulations indicated by the solid and dashed lines. The dashed red line is calculated using $\gamma = 3\times10^3 J\bullet m^{-3}\bullet K^{-1}$ ,while the solid red line is resulted from $\gamma = 6.2\times10^3 J\bullet m^{-3}\bullet K^{-1}$. (d) the simulated time evolution of electron temperatures

with various laser fluences.

**Figure 5. (color online). Atomistic simulation results for Co film.** (a) Ultrafast demagnetization curves with various microscopic damping values fitted by 3TM model as the solid lines. (b) The black dots represent the demagnetization time extracted from atomistic simulations as a function of microscopic damping constant and the exponential decay fitting is represented by the black line while the results obtained by Koopmans et al. as the red line.

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