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PAPER

Energy exchange dependent transient ferromagnetic like state of ultrafast magnetization dynamics

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Abstract

The study of laser-induced ultrafast magnetization dynamics is crucial for the development of information recording technology. Due to the complex mechanism, there is still a lack of comprehensive understanding for ultrafast magnetization dynamics. As an essential stage of laser-induced ultrafast magnetization switching process, the transient ferromagnetic like state (TFLS), has attracted much attention. Different from other studies on TFLS through the difference of magnetization dynamics between rare-earth and transition-metal, our study mainly focuses on the influence of energy injection and relaxation on TFLS in the process of ultrafast magnetization dynamics. The influence of various parameters on the formation of energy exchange dependent TFLS is studied. The results of simulation well support our view. Understanding the mechanism behind the TFLS is of great significance to promote the application of laser-induced ultrafast magnetization switching.

1. Introduction

Manipulating the direction of magnetization with laser pulse on picosecond or even femtosecond time scales is a very attractive technology which will have a major impact on the data storage industry. Since Beauripare first discovered ultrafast demagnetization in 1996 [1], there have been a lot of studies on laser-induced ultrafast magnetization dynamics [2–7], including helicity-dependent all-optical switching generated by the inverse Faraday effect [8, 9] and thermally induced magnetization switching which is considered to be the helicity-independent all-optical switching [10, 11]. However, the complete understanding of the complex mechanism of dynamics on this timescale is still unclear. The unexpected transient ferromagnetic like state (TFLS) resulting from the magnetization of rare-earth (RE) and transition-metal (TM) lattices paralleling to the same direction has attracted a lot of attention. This transient non-equilibrium state is considered to be a prerequisite for magnetization switching [12]. There have been many studies on the physics of TFLS formation [13–15], and it is believed that the difference of demagnetization rate between lattices is the main reason. For the ferrimagnetic material GdFeCo, experiments show that after the femtosecond laser pulse excitation, the lattices of FeCo and Gd exhibit different magnetization dynamics, that is, the TM reaches zero magnetization faster than RE. Then, with the lowering of the electron temperature, the magnetization of FeCo increases inversely and becomes parallel to the magnetization direction of Gd driven by the exchange field. This transient state, namely TFLS, lasts for several picoseconds and remains stable under the action of opposing magnetic fields [11]. It should be noted that TFLS is a strong transient parallel arrangement between the lattices, and it has been shown that the magnetic moments of FeCo and Gd can reach 25% of the equilibrium magnetization during this non-equilibrium state [14]. Besides GdFeCo, some studies indicate that TFLS can also occur in many other materials, such as TbFe alloys [16, 17]. This means that TFLS in laser-induced magnetization dynamics is a common phenomenon in these kinds of materials.

In this paper, different from other studies on TFLS through the difference of magnetization dynamics between lattices, we study the formation of TFLS from the perspective of energy exchange. The simulation results show that there is an energy threshold of laser pulse, resulting in the absence of TFLS. In this case, all the energy is used for demagnetization and the formation of TFLS requires additional energy of laser pulse. Further work shows that the energy injection, which is modulated by the fluence of laser pulse, initial temperature and damping is related to the duration of TFLS. Moreover, not only the energy injection but also the energy relaxation is an important factor for the formation of TFLS. In the simulations the relaxation of energy is modulated by the duration of laser pulse, as well as the electron–phonon coupling coefficient. The simulation results show that the relaxation of energy significantly affects the duration of TFLS. In general, our work indicates that the formation of TFLS is related to energy exchange. These findings help us to better understand the physical mechanism of ultrafast magnetization dynamics.

2. Magnetization dynamics model

To study the dynamics of TFLS under the excitation of femtosecond laser pulse, our simulations are based on the semiclassical spin model with atomic resolution [18], which is well adapted for describing the spin dynamics of GdFeCo [19]. The calculation of magnetization dynamics is based on a series of coupled Landau–Lifshitz–Gilbert (LLG) equations of motion. The energetics of the system is described by Hamiltonian, which is the Heisenberg model of the nearest neighbor spin exchange, given by:

$$\mathrm{H} = -\sum_{i\neq j} J_{ij} S_i \cdot S_j - \sum_i d_z S_{i,z}^2. \tag{1}$$

The Hamiltonian consists of near spin exchange and spin anisotropy. Where J_{ij} is the exchange integral between spin vector S_i and S_j (i, j are lattice sites), d_z is the uniaxial anisotropy constant (assumed along the Z axis). The spin moment in the formula $S_i = \mu_i/|\mu_i|$ is normalized, where μ_i is the magnetic moment of position i. In the LLG equation, the magnitude of the magnetic moment is fixed because only the transverse Gilbert damping is considered. For the ferromagnetic materials with the same spin direction, the adjacent spins are parallel, $J_{ij} > 0$; for the antiferromagnetic materials with antiparallel spin orientation, $J_{ij} < 0$. The LLG equation which is used to simulate magnetization dynamics, is described below:

$$\frac{\partial S_i}{\partial t} = -\frac{\gamma_i}{(1+\alpha_i^2)\mu_i} S_i \times (H_{\mathrm{eff}}^i + \alpha_i S_i \times H_{\mathrm{eff}}^i). \tag{2}$$

Here α_i and γ_i are Gilbert damping parameter and the gyromagnetic ratio respectively. H_{eff}^i is effective field. The right-hand side of the equation contains two parts. The first is the precession term, which describes the rotation of the spin under the action of the effective field. The second one is the damping term, which describes that the direction of spin inclines to the effective field direction gradually due to the influence of the effective field. H_{eff}^i contains the deterministic Hamiltonian part and thermal noise, as described in the following equation:

$$H_{\mathrm{eff}}^i = -\frac{\partial \mathrm{H}_i}{\partial S_i} + \zeta_i. \tag{3}$$

Here ζ_i represents a stochastic term, which describes the coupling with the external heat bath [20]. As a part of the effective field, the thermal fluctuations are not related to time and space. The correlators of different components of this field can be written as:

$$\langle \zeta_{i,a}(\mathrm{t}) \rangle = 0 \tag{4}$$

$$\langle \zeta_{i,a}(t)\zeta_{i,b}(t') \rangle = \frac{2\mu_i k_{\mathrm{B}}}{\gamma_i} \alpha_i T \delta_{ij} \delta_{ab} \delta(t - t'), \tag{5}$$

where a and b refer to the Cartesian components of the spin vector, and i and j refer to separate spins (i.e., spatially unrelated). T is the temperature of heat bath to which the spin is coupled. α_i is the damping parameter, which describes the efficiency of energy exchange between laser pulse and magnetic system. Since the amount of Co is small and both Fe and Co are coupled ferromagnetically, for simplicity we focus on the magnetization behavior of Fe and Gd, ignoring Co. The GdFe alloy system is filled with 32 × 32 × 128 unit cells. The face center cubic lattice is filled with TM and RE lattices in the desired concentration 25% and 75%, respectively. The effective magnetic moments for the Fe and Gd sublattice are $\mu_{\mathrm{Fe}} = 1.92\ \mu_{\mathrm{B}}$ and $\mu_{\mathrm{Gd}} = 7.63\ \mu_{\mathrm{B}}$ respectively, where μ_{B} is the Bohr magneton.

Table 1. Values of the parameters in our simulation.

Quantity	Value	Units
k_B (Boltzmann constant)	1.38×10^{-23}	$J \cdot K^{-1}$
μ_B (Bohr magneton)	9.27×10^{-24}	$A \cdot m^2$
Γ (gyromagnetic ratio)	1.76×10^{11}	$T^{-1} \cdot s^{-1}$
α (Gilbert damping parameter)	Varied	
G_{ep} (electron–phonon coupling factor)	Varied	
C_p (phonon specific heat capacity)	3×10^6	$J \cdot m^{-3} \cdot K^{-1}$
γ_e (electronic specific heat constant)	7×10^2	$J \cdot m^{-3} \cdot K^{-2}$

To better describe the effect of the thermal field generated by the laser pulse on the magnetic system, we employ the two-temperature model [21]. The model consists of two heat baths, conducting electrons and phonons. The energy of the laser pulse is absorbed by the conducting electrons first, and then relaxed into the phonons through the electron–phonon coupling [22]. The two-temperature model can be used to describe the temperature changes of electrons and phonons in this process. The coupled differential equations of temperature dynamics are described as follows:

$$C_e(T_e)\frac{dT_e}{dt} = -G_{ep}\left(T_p - T_e\right) + P(t) \tag{6}$$

$$C_p\frac{dT_p}{dt} = -G_{ep}\left(T_e - T_p\right), \tag{7}$$

where $C_e(T_e) = \gamma_e T_e$ and γ_e is electronic specific heat constant [23]. C_p is phonon specific heat capacity. G_{ep} is electron–phonon coupling factor [10]. In our work we assume that the electronic temperature T_e is coupled to the magnetic system through equation (5) [10]. $P(t)$ in the formula is the power of laser pulse varying with time, which is related to the fluence of laser pulse P_0 and the pulse duration τ_p, given by:

$$P(t) = P_0 \exp(-((t - \tau_p)/\tau_p)^2). \tag{8}$$

The detailed steps and reliability of numerical simulation in this paper were discussed in our previous work [10, 18, 24]. The values of parameters are shown in table 1:

3. Results and discussion

The process of laser-induced ultrafast magnetization dynamics is shown in figure 1. Inset of figure 1 shows the time dependence of the electron and phonon temperature from the two-temperature model. It demonstrates clearly that the excitation of the laser pulse makes the energy rapidly absorbed by the electron thermal bath, resulting in a sharp rise of electron temperature, which reaches the maximum value in less than 1 ps.

The result of the rapid rise of electron temperature is that the Langevin field (thermal excitation field) overcomes the exchange field and makes the independent demagnetization of Fe and Gd lattice. Different from 3d ferromagnets, the magnetism of Gd is dominated by 4f electrons which cannot be excited directly by laser pulse [25], resulting in the demagnetization of Gd slower than that of Fe. The exchange field dominates again after the electron temperature cools down, and the demagnetization of Gd leads to the reverse magnetization of Fe due to angular momentum conservation. At this time, there is a short strong non-equilibrium state in which the magnetization direction of Fe and Gd are in the same direction, as shown in the ellipse area of figure 1, which is called TFLS. When Gd is completely demagnetized, TFLS ends.

The above content explains the formation of TFLS from the perspective of the difference of magnetization dynamics between Fe and Gd. In this paper, we attempt to analyze the formation of TFLS from the energy exchange of magnetic system. The ability to reverse the state of the magnetization is thought to require a directional stimulus to break the symmetry of the system. During the ultrafast magnetization dynamics, the laser pulse puts thermal energy at a certain rate into the electron system, causing dramatic overheating of the electron. Such overheating is crucial for demagnetization, which leads to ultrafast demagnetization of Fe and Gd. When the laser energy is just above certain threshold (sufficient energy to switch but not to form the no-equilibrium TFLS), magnetization switches without the formation of TFLS. In other words although the laser pulse provides sufficient energy to reverse the magnetization but no surplus to cause the sublattices to become parallel. If the energy provided is then increased, any extra energy can then lead to the TFLS. According to the two-temperature model, the injection of energy makes overheating of the electron (sufficient extra energy). This stimulus broke the balance of the magnetic

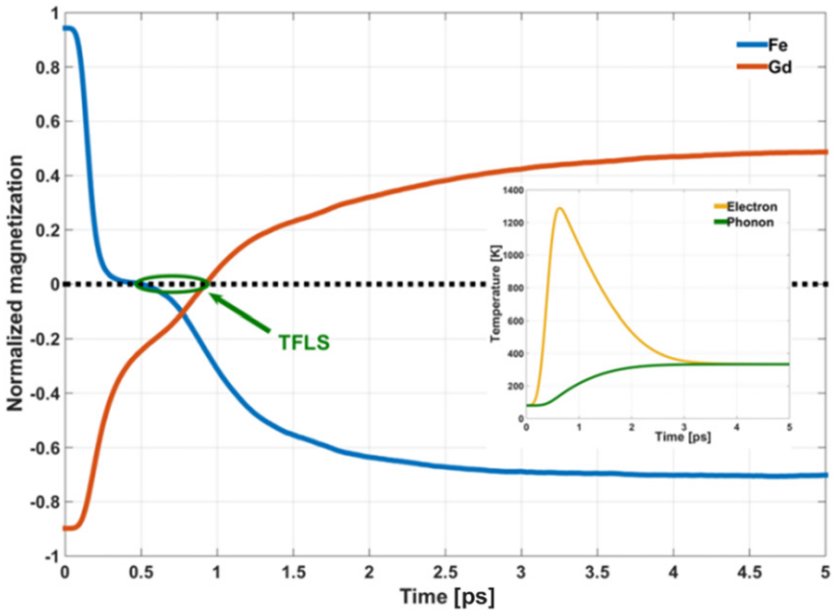

Figure 1. Diagram of laser-induced ultrafast magnetization dynamics. The blue line and red line represent the magnetization evolution of Fe and Gd, respectively. In the ellipse area, the magnetization of Fe and Gd are in the same direction, which represents TFLS. (Inset) Time dependence of the electron and phonon temperature from the two-temperature model. The yellow line and green line represent the temperature evolution of electron and phonon, respectively.

system, leading to the formation of TFLS. Meanwhile, if the injection of energy cannot be completed in a short time, it will lead to more energy relaxation into the phonon. This makes the overheating of the electron (the extra energy) unable to maintain a high value and causes the duration of TFLS to become shorter. Hence the injection and relaxation of energy are both important factors, which determine the duration of TFLS. The influence of various parameters on the formation of energy dependent TFLS are discussed in the following part.

Firstly, relationship between the fluence of laser pulse and the duration of TFLS is studied. The data points in figure 2 describe durations of TFLS excited by laser pulse with different fluences. It can be clearly seen that the increase of the fluence of laser pulse leads to more energy injection, which significantly prolongs the duration of TFLS. By fitting the data points in the figure and extending the fitting line in reverse, it can be found that the fitting line intersects with the coordinate axis near 0.32 GJ m^{-3}. With this amount of excitation fluence, the corresponding duration of TFLS is equal to zero. This means that there is a threshold fluence. When the laser pulse with the threshold fluence excites the sample, the energy only meets the magnetization switching. If the fluence of laser pulse continues to increase, the extra energy in the electron system needs a finite time to be relaxed, which leads to the formation of the highly non-equilibrium state of TFLS. The simulation results show that the formation of TFLS is strongly dependent on the injection energy. In addition, other studies have shown that the increase of pulse fluence leads to faster demagnetization [26]. Dominated by different electrons, the increase of pulse fluence has greater effect on the demagnetization of Fe than that of Gd. The results lead to a larger difference in demagnetization between the lattices and longer TFLS, which validates our simulation.

In the following part the influence of initial temperatures on the duration of TFLS is studied. In fact, the effect of the initial temperature and the fluence of laser pulse on the magnetic system are very similar in that they both increase the injection of energy into the magnetic system. The increase of initial temperature can directly lead to the increase of electron temperature and phonon temperature in the first place. In contrast, increasing the fluence of laser pulse only significantly increases the electron temperature, but the increase of phonon temperature mainly depends on the electron–phonon interaction. As shown in figure 3, the simulation results show that the increase of the initial temperature makes the duration of TFLS longer. The reason is similar to that of the fluence of laser pulse discussed above. The increase of initial temperature injects more energy into the magnetic system at the beginning. After driving ultrafast demagnetization, the extra energy needs more time to relax into the phonon, resulting in the formation of TFLS with longer duration. In addition, the simulation are repeated with different fluences of laser pulse. It can be seen from figure 3 that different lines show a consistent trend, the duration of TFLS increases with the increase of initial temperature, which indicates that the conclusion of the relationship between initial temperature and the duration of TFLS is universal.

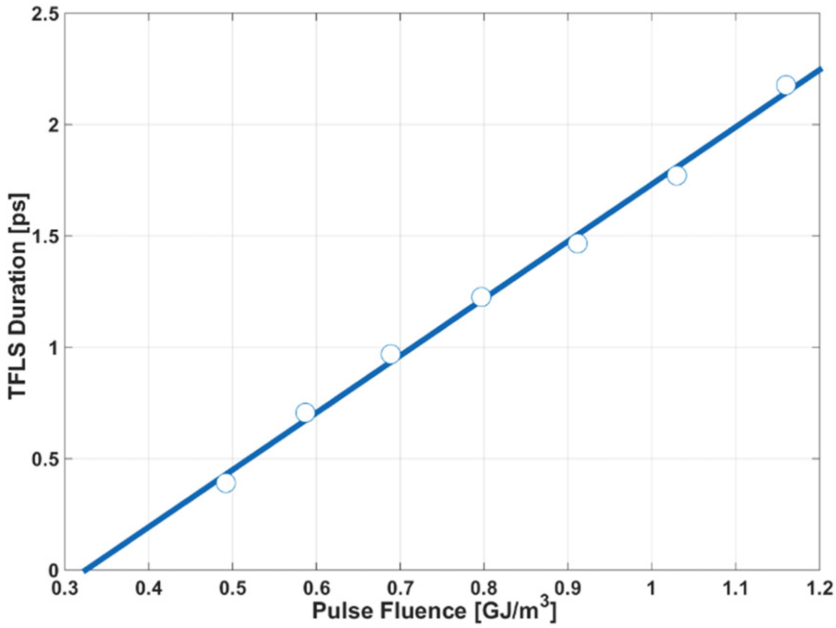

Figure 2. Relationship between the fluence of laser pulse and the duration of TFLS. The circles represent the duration of TFLS under different fluence, and the line is fitted to illustrate the evolution of duration of TFLS.

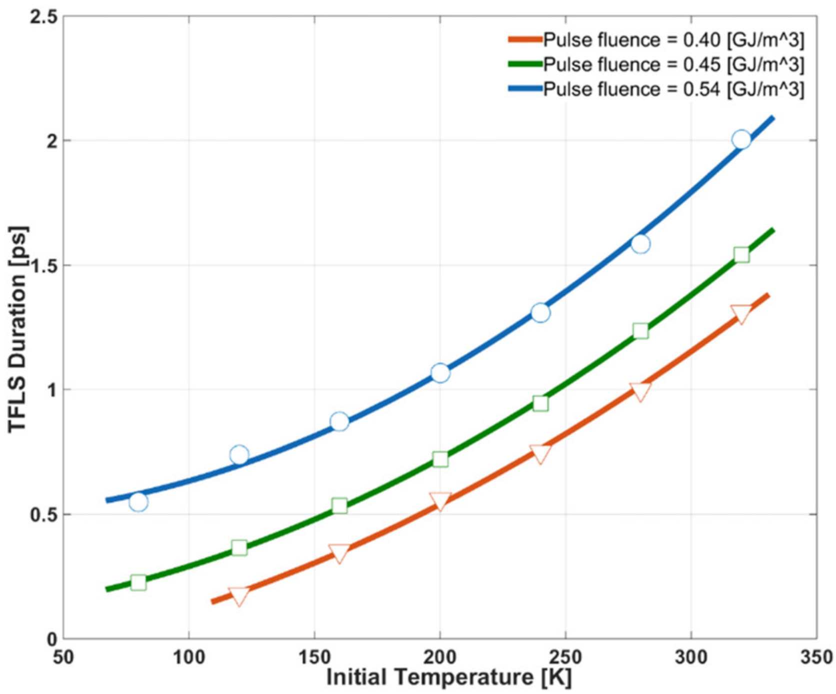

Figure 3. Relationship between initial temperature and TFLS. Data points represent the duration of TFLS at different initial temperatures. Data points represent the duration of TFLS at different initial temperature. Different colored lines represent different fluences of laser pulse.

In the first two parts, our research focuses on the control of the amount of energy entering the magnetic system, such as changing the fluence of laser pulse and the initial temperature. While the energy is absorbed by electrons, part of it is relaxed into the phonon. If the energy of laser pulse cannot enter the system in a short time, the relaxed energy becomes a non-negligible factor. In this part, the rate of energy entering the magnetic system is modulated by changing the pulse duration. The simulation results are shown in figure 4. It can be seen that for the laser pulse with the same fluence, increasing the duration of laser pulse shortens the duration of TFLS. The increase of the duration of laser pulse slows down the rate of the energy entering the system. At the same time, the coupling between electron and phonon relaxes part of the energy, so that

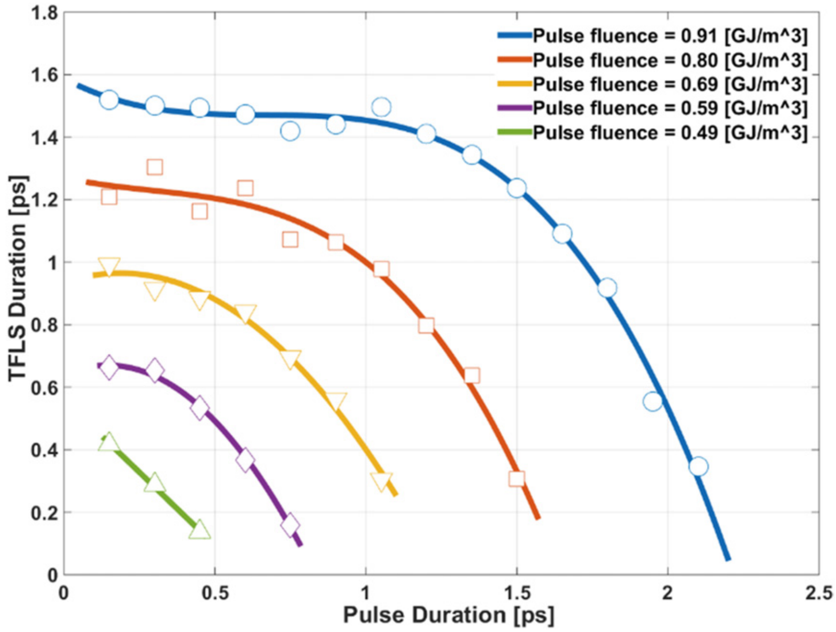

Figure 4. Relationship between the duration of laser pulse and the duration of TFLS. The data points represent the duration of TFLS under laser pulse with different duration. Data points represent the duration of TFLS at different pulse duration. Different colored lines represent different fluences of laser pulse.

the amount of extra energy forming TFLS cannot be maintained at a high value. As a result, the duration of TFLS becomes shorter. The results also validate the previous work of Davies *et al* [27], whose results indicate that increasing the pulse duration leads to increasingly comparable demagnetizing rates of Gd and Fe. In addition, the simulation are repeated with different fluences of laser pulse. The results show that the TFLS caused by laser pulses with different fluences are not the same. It can be found that the magnetic system excited by laser pulse with small fluence is more sensitive to the duration of laser pulse, where the change of TFLS is strongly related to the duration of laser pulse. By contrast, for the magnetic system excited by laser pulse with large fluence, the increase of the duration of laser pulse has little effect on TFLS at the beginning. Only when the duration of laser pulse becomes large enough will the TFLS become significantly shorter. These simulations indicate that there is a threshold for the amount of energy. Above this specific value, the change of energy will not have a significant impact on TFLS. On the contrary, when it is reduced below the threshold, the change of energy will significantly affect the duration of TFLS.

As mentioned before, the amount of the energy injection and the rate of energy relaxation are closely related to the duration of TFLS. Not only the external parameters, but also the internal parameters can control the energy exchange. In this part, the same laser pulses are used to excite magnetic systems with different damping. The simulation results are shown in figure 5. It should be noted that the horizontal axis represents the ratio of the Fe damping α_{Fe} to the Gd damping α_{Gd}, and the different lines represent different values of Fe damping. As shown in figure 5, the duration of TFLS decreases with the increase of the ratio $\alpha_{\mathrm{Fe}}/\alpha_{\mathrm{Gd}}$ (the decrease of Gd damping). Damping is a very important parameter, which is considered to be the external manifestation of the interaction of various internal factors in the magnetic system [28, 29]. Spin orbit coupling is considered to be the main internal source of damping [30]. This means that the value of damping represents the rate of spin reaching equilibrium. When the same energy of laser pulse excites the magnetic system, the smaller Gd damping will slow down the rate of spin reaching equilibrium, resulting in more energy required for demagnetization. Therefore, there is not enough extra energy to form the TFLS with long duration. Furthermore, other damping cases are simulated, as shown by the lines of other colors in figure 5, and the simulation results present consistency.

The internal parameters of the magnetic system can also affect the relaxation of the energy. Electron phonon coupling coefficient G_{ep} is a key parameter to control the energy relaxation rate between electron and phonon. The larger the G_{ep}, the higher the energy transfer efficiency between the electron and the phonon. The same laser pulses are used to excite the magnetic systems with different G_{ep}. The simulation results are shown in figure 6, indicating that the TFLS with larger G_{ep} has a shorter duration. When a fixed energy is injected into the magnetic system, due to the increase of G_{ep}, the relaxation of energy from electron to phonon will be accelerated. As a result, the extra energy used to support the presence of a highly non-equilibrium TFLS will decrease. Furthermore, the conclusion that the duration of TFLS decreases with

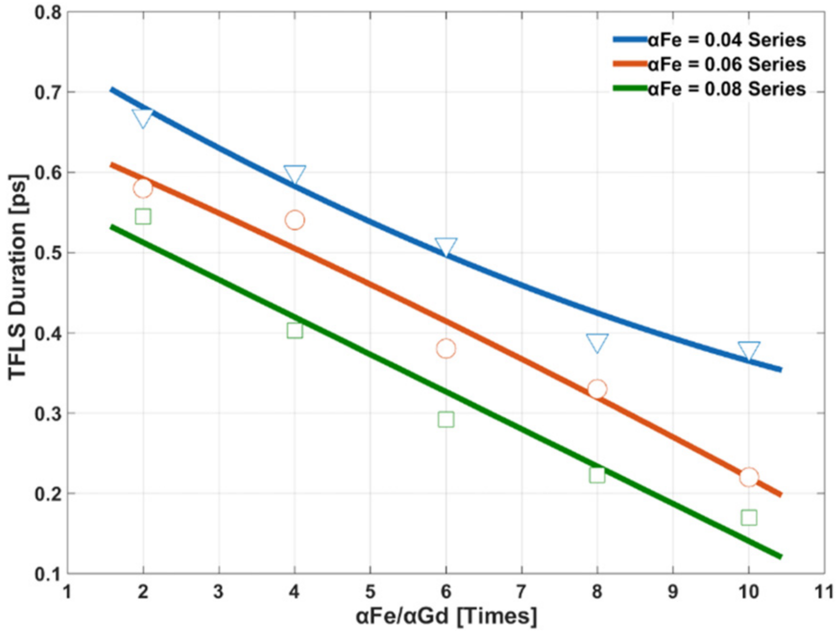

Figure 5. Relationship between damping and the duration of TFLS. The horizontal axis represents the ratio of Fe damping to Gd damping. Data points represent the duration of TFLS at different damping ratio. The colors of the lines represent different damping values.

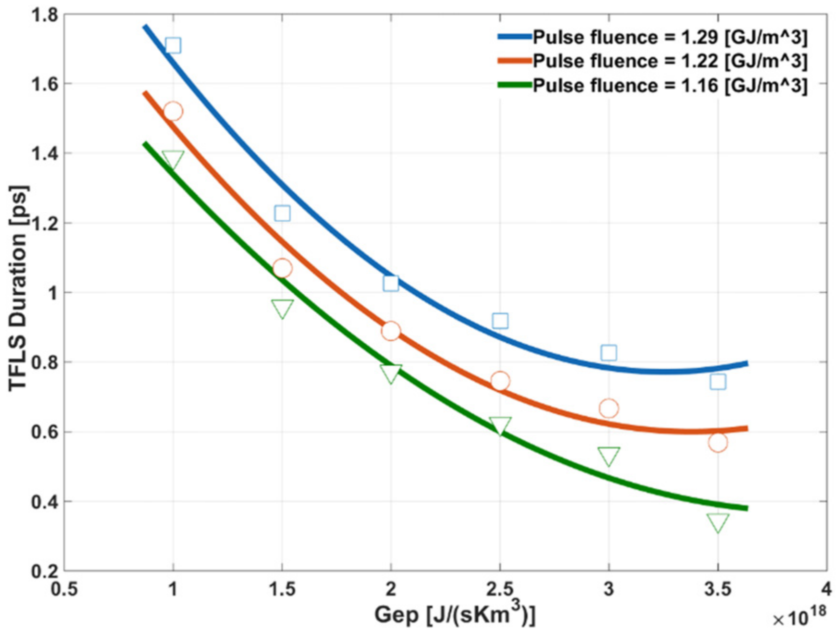

Figure 6. Relationship between electron phonon coupling coefficient G_{ep} and TFLS. Data points represent the duration of TFLS with different G_{ep}. Different colored lines represent laser pulse with different fluences.

the increase of G_{ep} is still valid under the excitation of laser pulse with different fluences, as shown in figure 6.

4. Conclusions

As an essential stage of ultrafast magnetization dynamics, TFLS connects demagnetization and magnetization switching. The essence of TFLS is ascribed to the difference of demagnetization rate between Fe and Gd. Specifically, the magnetization direction of Fe lattice is the same as that of Gd lattice, resulting in a transient non-equilibrium state. In this paper, we demonstrate that the formation of TFLS is related to the energy exchange. Our study mainly focuses on how the injection and relaxation of energy affect the TFLS.

To that end, the effects of various parameters on the energy exchange are simulated. It is found that the increase of fluence of laser pulse and initial temperature increase the amount of energy injection and lengthen the duration of TFLS. The rate of energy injecting into the magnetic system can be slowed down by increasing the pulse duration, which allows more energy to be relaxed, leading to a shorter duration of the TFLS. Furthermore, the parameters of the material, namely, the damping and the electron phonon coupling coefficient G_{ep} are also studied. The simulation results show that the smaller damping can slow down the rate of spin reaching equilibrium, resulting in more energy required for demagnetization. Therefore, the duration of TFLS which is formed by extra energy will be shorter. In addition, the simulation results show that G_{ep} changes the rate of energy relaxation to the phonon, which affects the duration of TFLS. Unlike previous studies on TFLS from the perspective of differences of magnetization dynamics between RE and TM, here we propose a view that TFLS is related to energy exchange, which are well supported by the results of our simulations. This view helps us better understand the formation of TFLS, revealing that the mechanism of TFLS is of great significance for laser-induced ultrafast magnetization switching.

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Data availability statement

The data generated and/or analysed during the current study are not publicly available for legal/ethical reasons but are available from the corresponding author on reasonable request.

Conflict of interest

The authors declare no conflicts of interest.

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