Ultrafast reduction in exchange interaction by a laser pulse: alternative path to femtomagnetism

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Abstract
Since the beginning of femtomagnetism, it has been hotly debated how an ultrafast laser pulse can demagnetize a sample and switch its spins within a few hundred femtoseconds, but no consensus has been reached. In this paper, we propose that an ultrafast reduction in the exchange interaction by a femtosecond laser pulse is mainly responsible for demagnetization and spin switching. The key physics is that the dipole selection rule demands two distinctive electron configurations for the ground and excited states and consequently changes the exchange interaction. Although the exchange interaction change is almost instantaneous, its effect on the spin is delayed by the finite spin wave propagation. Consistent with the experimental observation, the delay becomes longer with a stronger exchange interaction pulse. In spin-frustrated systems, the effect of the exchange interaction change is even more dramatic, where the spin can be directly switched from one direction to the other. Therefore, our theory has the potential to explain the essence of major observations in rare-earth transition metal compounds for the last seven years. Our findings are likely to motivate further research in the quest of the origin of femtomagnetism.

Keywords: exchange interaction, femtomagnetism, Heisenberg model

1. Introduction
In magnetism, exchange interactions among electrons play a central role in sustaining a long-range magnetic ordering across lattice sites [1–17]. If the exchange interaction is suddenly quenched, the consequence would be catastrophic, where the spins could be decoupled and a magnet could suddenly become nonmagnetic. In femtomagnetism [18–22], a laser field excites electrons out of the Fermi sea and alters the electron configuration and electron-electron interaction (Coulomb and exchange interactions, see figure 1), whose change further affects the excitation of the electrons. Since any pair of electron excitations must obey the dipole selection rule rigorously, it is conceivable that the effect must be quite strong. So far, experimental evidence for the exchange interaction change has been scant, mostly for the exchange splitting change in the photoemission spectrum [23, 24].

For the Gd(0001) surface states, Lisowski \textit{et al} [25] showed that while the spin polarization is reduced by half, the exchange splitting is unchanged. A more recent experiment in gadolinium [26] shows that the exchange splitting is reduced by 0.2 eV, where the binding energy increases by 0.1 eV for the minority band and decreases for the majority band by the same amount. In Gd\textsubscript{0.55}Sr\textsubscript{0.45}MnO\textsubscript{3}, Matsubara \textit{et al} [27] showed that it is possible to switch on the double exchange interaction between ions and in Eu\textsubscript{1−x}Gd\textsubscript{x}O they [28] found that depending on the doping level of Gd, the exchange interaction can be increased or decreased. Up to now, it is unclear whether the exchange change is the cause or the result of the demagnetization, but as pointed out by Bigot \textit{et al} [29],
there is little doubt that the exchange interaction plays a crucial role here.

Theoretically, the exchange interaction may be temperature dependent [30], but there has been no such study to directly address the exchange interaction change due to the laser excitation in the time domain. The temperature concept is invalid on the femtosecond time scale. In our first theoretical attempt, we manually changed the exchange interaction strength [31] and found that the reduction of the exchange interaction prolongs the spin relaxation, which was confirmed experimentally [32]. Stamenova and Sanvito [33] only addressed the dynamical exchange interaction due to a magnetic field, but their dynamic exchange interaction is still time-independent since they time-average the exchange coupling parameter. Therefore, a theoretical investigation is very appropriate and important, not only for femtomagnetism, but also for other research fields.

For instance, ultrafast laser pulses have long been extensively used to investigate the origin of the high-temperature superconductivity over two decades [34–39]. Since the electron correlation and the electron–phonon coupling act on different time scales, one can separate them in the time domain. Similarly, magnetic and nonmagnetic contributions can be separated as well. By weakening the exchange interaction, one can suppress the antiferromagnetic ordering and enhance the non-magnetic contribution. This allows one to isolate each effect separately to examine whether or how the antiferromagnetic coupling is essential to Cooper pairing in high-temperature superconductors.

In this paper, we theoretically show that a laser pulse is able to change the exchange interaction dynamically; and as a result, the spin dynamics (demagnetization and spin switching) is strongly affected. Due to the optical selection rule, the excited-state configuration must differ from the ground state, which changes the exchange interaction. The amount of change depends on the field amplitude and the helicity of the light, where a much larger change is found for circularly polarized light. For a pure ferromagnetic and antiferromagnetic coupling, for the first time we demonstrate a delayed response of the spin system with respect to the exchange pulse and the delay becomes longer with a smaller exchange interaction, consistent with the experimental observations. In spin-frustrated systems, such as rare-earth transition metal compounds, the change is even more dramatic. For each of three different spin configurations investigated here, we find that the exchange pulse can change the course of spin dynamics. Importantly, the spins do not feel the exchange pulse immediately; instead, after several hundred femtoseconds when the exchange pulse is over, the main spin change starts, similar to the experimental results in GdFeCo [40]. By changing the exchange pulse amplitude, we are able to show that it is indeed possible that a small change in the exchange amplitude can lead to dramatically different spin dynamics, but this is only possible in spin-frustrated systems and when the pulse amplitude is strong enough. This provides a new clue as to why experimentally changing the laser intensity by little as 0.05 mJ cm\(^{-2}\) could lead to the helicity-independent switching, a nonthermal all-optical switching. Therefore, we strongly believe that the exchange interaction change is an alternative path to femtomagnetism.

This paper is arranged as follows. In section 2, we explain how the exchange interaction is changed during the laser excitation. Section 3 is devoted to the effect of the exchange interaction pulse on the spin dynamics and switching. In section 4, we apply our theoretical results to experiments. Finally, we conclude the paper in section 5.

2. Exchange interaction change in excited states

To illustrate how the exchange interaction is changed during the photoexcitation, we consider a transition from the ground state \(\psi_{\text{gs}} = (\phi_a(1)\phi_b(2) - \phi_c(1)\phi_d(1))/\sqrt{2}\) to the excited state \(\psi_{\text{ex}} = (\phi_a(1)\phi_b(2) - \phi_c(1)\phi_d(1))/\sqrt{2}\), where all the \(\phi\) are single-particle wavefunctions. The exchange integrals in the ground and excited states are \(J_{\text{gs}} = -\langle \phi_a \phi_b | r_{12}^{-1} | \phi_c \phi_d \rangle\) and \(J_{\text{ex}} = -\langle \phi_a \phi_d | r_{12}^{-1} | \phi_c \phi_b \rangle\), respectively, where the Dirac brackets represent the double integrations over the coordinates of two electrons and \(r_{12}\) is the distance between two electrons. Here we do not explicitly distinguish the difference between the exchange interaction and exchange integral, since the topic has been discussed many times in the literature [2–16]. Usually, these two exchange integrals are independent of each other, but optical transitions require that the exchange integral in the excited state has only four independent terms: 

\[
-\langle \phi_a \phi_b | r_{12}^{-1} | \phi_c \phi_d \rangle, -\langle \phi_a \phi_d | r_{12}^{-1} | \phi_c \phi_b \rangle, -\langle \phi_b \phi_d | r_{12}^{-1} | \phi_a \phi_c \rangle, -\langle \phi_b \phi_c | r_{12}^{-1} | \phi_a \phi_d \rangle \]

(see the appendix A for details). Since the excited state is much more delocalized, the exchange interaction is in general smaller.

We can directly evaluate those exchange integrals. Consider two electrons on two lattice sites separated by a distance \(R\). The ground-state wavefunction is chosen to be the Slater type 3d\(\sigma\) orbitals localized at two different sites, while the excited-state wavefunction has one electron in the 3d\(\sigma\) orbital and the other promoted to the 4p\(\sigma\) orbital. This particular choice of the electron configuration resembles the 3d transition metals, excited with a linearly polarized light. All the exchange integrals are computed using the Deric program [41, 42], with the orbital exponents \(\xi\) and orbital quantum numbers as the input parameters shown in the figure [43, 44]. Figure 2(a) shows that \(J_{\text{gs}}(3d\sigma, 3d\sigma | 3d\sigma, 3d\sigma)\) is in general much larger than \(J_{\text{ex}}(3d\sigma, 4p\sigma | 3d\sigma, 4p\sigma)\) for most of the distances.
For the rare-earth compounds, the ground state consists of the 4f orbital and the excited state has one electron promoted to the 5d orbital. Figure 2(b) shows the same trend for the exchange integral. However, it has been observed experimentally that the linearly and circularly polarized lights have different thresholds for the nonthermal all-optical writing. We also compute the exchange interaction change for circularly polarized light. Our finding is very interesting. Irrespective of whether 3d or 4f orbitals are excited, the circularly polarized light reduces the exchange integral much more. The solid lines in figures 2(c) and (d) refer to the results obtained with the linearly polarized light, while the dashed lines denote the exchange integrals obtained with the circularly polarized light. The difference is very clear. The effect in 4f is more pronounced, where the exchange interaction is reduced close to four-fold. The reason for the reduction is because the circularly polarized light only excites π orbitals. Spatially, there is a very small overlap with σ orbitals, so the exchange integral becomes smaller. This explains why experimentally the circularly polarized light is more effective than linearly polarized lights.

As seen from the above discussion, the exchange interaction change depends on a specific pair of states. Experimentally, a laser pulse has a narrow photon energy region that naturally select a few transitions. With a longer laser pulse, the energy window is narrower. The band structure of a material further limits which states participate. For the 3d transition metals, the transition is mostly from occupied 3d states to unoccupied hybridized 4p states. This means that even for a collective excitation where multiple states participate, the transitions have the same character. Our prior study shows that in fcc Ni, along those high symmetry lines, only two dominate the initial excitation [45]. However, for more complicated materials, it is necessary to evaluate the exchange interaction change individually, by taking into account both the extrinsic (laser field) and intrinsic (material) parameters.

3. Effects of dynamic exchange interaction on spin dynamics and switching

Once a laser impinges on a magnetic sample, the laser pulse excites the electronic systems and changes the exchange interaction. Thus, the laser effectively introduces a time-dependent exchange interaction pulse [46],

\[ J(t) = J_0 \left(1 - Ae^{-(t-T)/\tau}\right), \]

where \( J_0 \) is the static exchange interaction, \( T \) is the time delay of the exchange pulse and \( \tau \) is the duration time of the pulse, typically on the order of 20–100 fs. \( A \) is the dimensionless amplitude changing from 0 (no laser excitation) to 1 (complete quenching of the exchange interaction). To be definitive, we choose \( J_0 = 0.02 \text{ eV} \), but one has to keep in mind that those

![Exchange integral as a function of the distance between two atoms.](image-url)
exchange interaction parameters may differ a lot for rare-earth compounds. In this study, we have made no attempt to fine-tune them or distinguish them among different ions, since there are lots of excellent research done previously in this field (see, for instance [47, 48]). Our envelope function is an inverted Gaussian and ensures that in both the negative and positive infinite times, the system has the same $J_0$. The exchange drops only during the narrow time window determined by the duration. In realistic excitations, the pulse may not be symmetric in the time domain and may have a long tail; physically the excited states may live quite a bit longer than the laser excitation, or the optically excited states might thermalize into other excited states. As a first step, we assume that the exchange can be changed only when the electrons transit to the excited states. This assumption allows us to investigate whether such a shortest exchange pulse has a lasting impact on the spin dynamics.

In the following, we consider two systems: one-dimensional spin chain and a cube of eight spins. It should be pointed out that due to the Mermin–Wagner theorem [49], at any non-zero temperature, one or two-dimensional isotropic Heisenberg systems can not be either ferromagnetic or antiferromagnetic. For this reason, our system is always kept at zero temperature, so the magnetic ordering is always maintained.

### 3.1. One-dimensional spin chain

To appreciate how the exchange interaction change affects the spin dynamics, we choose a chain of $N = 40$ spins coupled through the Heisenberg exchange interaction,

$$H = -\sum_{ij} J_{ij}(t) \mathbf{S}_i \cdot \mathbf{S}_j, \quad (2)$$

where $\mathbf{S}_i$ is the spin operator at site $i$ and the summation is over the nearest-neighbor sites only. This Hamiltonian differs from the commonly used Heisenberg model by the time-dependent exchange interaction [50]. In principle, $J_{ij}$ should be different for different ions. Wienholdt and coworkers [47] even proposed the orbital-resolved exchange interaction. Our focus is on the qualitative understanding of the ultrafast magnetization switching among different materials, since there have been extensive investigations in the literature [51] and even the software package is available [52]. We adopt the semi-classical method that treats the spin as a unit vector. The dynamic evolution follows the equation of motion

$$\frac{d \mathbf{S}_i}{dt} = \sum_{ij} J_{ij}(t) \mathbf{S}_i \times \mathbf{S}_j, \quad (3)$$

where the summation is over the nearest neighbors only. Since the exchange change is almost instantaneous, the spin can feel the time-dependence of the exchange’s change only when the electron is excited.

We investigate three different magnetic orderings (see figure 3): ferromagnetic, antiferromagnetic and spiral orderings. We start with a ferromagnetic configuration with all the spins along the $z$-direction initially. We note in passing that for pure ferromagnetic and antiferromagnetic configurations, there is no torque exerted on the spins and an initial tilting is necessary to start the spin dynamics. For all the other configurations, this step is unnecessary. To start the dynamics, at $t = 0$ we tilt the spin at site $i = 1$ by $10^\circ$ from the $z$-axis to the $y$-axis and allow the spins to precess by themselves for some period of time, since experimentally the spin precession is always present even before the laser pulse. At $T = 200$ fs, we turn on the exchange pulse of duration $\tau = 40$ fs and amplitude $A = 0.5$. This 200 fs is referenced with respect to the initial time $t = 0$. If the initial time is shifted to a new time, one has to shift the peak time as well. Figure 4(a) shows that the spin at site $(i = 30)$ $S_{30y}$ precesses with time in the absence (dotted line) and in the presence (solid line) of the exchange pulse. We choose the $x$-component of site $i = 30$ because it is representative enough as it is far away from the first site.

Our results show that the spin dynamics with the exchange pulse is delayed by 37 fs with respect to that without the pulse. The absolute delay depends on (a) the geometrical distance between the excitation site (whose exchange interaction is changed) and the location of observation and (b) the nature of the magnetic ordering (see below). We also compute the spin dynamics with $T = 700$ fs, where the results are similar (see the vertically shifted thin line in figure 4(a)). The overall spin precession is shifted to a longer time scale, but the details of the dynamics remain the same. This is consistent with experimental observations in 3d transition metals [53, 54]. The reason for this delay can be seen directly from equation (3). Once the exchange is reduced, the torque on the spin is smaller and consequently the spin dynamics slows down. This leads to a linear dependence of the delay on the pulse duration $\tau$. The longer the pulse is, the longer the delay becomes (see the inset in figure 4(a) with $T = 200$ fs). This conclusion remains the same for the antiferromagnetic ordering and the spiral ordering (results are not shown). Other than this time delay, the spin dynamics remains the same, but what if not all the sites have the same exchange change?

Experimentally, only a small portion of the sample is illuminated by the laser pulse. We wonder whether this uneven
configuration. The solid line represents the $x$-component of spin $S_{30}$, at site 30 with the exchange pulse; the dotted line denotes the spin precession without the exchange pulse; and the thin line is the spin change for $T = 700$ fs (vertically shifted for clarity). Inset: dependence of the first spin peak time on the exchange pulse duration $\tau$ with $T = 200$ fs. Here the exchange pulse is applied uniformly to all the sites. (b) Spin precession as a function of time for the spiral configuration. A dramatic change is seen with the exchange pulse applied on sites between 20 to 30. The thin solid line is the exchange pulse.

Figure 4. (a) Spin precession with time for the ferromagnetic configuration. The solid line represents the $x$-component of spin $S_{30}$ at site 30 with the exchange pulse; the dotted line denotes the spin precession without the exchange pulse; and the thin line is the exchange pulse. A dramatic change is seen with the exchange pulse applied on sites between 20 to 30. We focus on the $x$-component of spin $S_{30}$, at site 30 with the exchange pulse; the dotted line denotes the spin change for $T = 700$ fs (vertically shifted for clarity). Inset: dependence of the first spin peak time on the exchange pulse duration $\tau$ with $T = 200$ fs. Here the exchange pulse is applied uniformly to all the sites. (b) Spin precession as a function of time for the spiral configuration. A dramatic change is seen with the exchange pulse applied on sites between 20 to 30. The thin solid line is the exchange pulse.

Figure 5. Five spin configurations on a cube. The site number convention is given on the top left cube. No. 804 is ferromagnetic, No. 806 is antiferromagnetic, No. 801 is an in-plane canted spin configuration, No. 805 is antiferromagnetic in-plane while ferromagnetic inter-plane and No. 802 has all the nearest neighboring spins perpendicular to each other. These initial configurations are used in the real simulation.

3.2. A simple cube of eight spins

Up to now, our focus has been on the linear spin chain, where the spin wave propagates from one site to the next. It is natural to ask what happens in higher dimensions. Due to the high complexity, we restrict ourselves to a cube of eight spins and five different spin orderings (see figure 5), with a focus on the main physics. For larger systems, the reader is referred to some of the latest publications [47]. We find that despite the diverse magnetic orderings, if the exchange interaction pulse acts upon all the lattice sites uniformly and simultaneously, the spin change is also delayed. Following the above investigation, we consider the cases where the exchange interaction pulse is only applied upon to sites 1 and 2 (see configuration No. 804\(^{3}\) in figure 5 for the numbering convention), while all the other sites have a constant exchange interaction $J_p$. This approach emphasizes the interaction change between different spin sites. In our system, we have different spin configurations between nearest-neighbor sites, where our interest is on the femtosecond dynamics. Experimentally, GdFeCo samples are amorphous and exhibit laser-induced ultrafast switching. Building some inhomogeneity into our model simulates realistic experimental situations better.

With the above consideration in mind, we start to explore the dynamics in five structures. For FM (No. 804), we find that the overall change due to the exchange pulse is small (the results are not shown). This is similar to the linear spin chain. For AFM (No. 806), in figure 6(a) we show how the $x$-component of spin $S_{7}$ at site 7 changes with time. This site is chosen since it exhibits a much more pronounced change. We notice that without the exchange pulse, $S_{7}$, gradually increases with time from 0 fs to 600 fs, with a very small oscillation (see the dotted line). When we turn on the exchange pulse at sites 1 and 2 (see the thin solid line), the oscillation becomes stronger.

\(^{3}\) The configuration number is only for our programming purpose to distinguish various configuration. It has no special meaning.
The delay between the exchange pulse and spin dynamics has important consequences for experiments. Theoretically, this delay is expected since the spin wave needs time to propagate from one site to the next. For a normal uniform magnetic ordering such as FM and AFM, the collective spin excitation can survive the exchange interaction change. Physically, they have a larger spin stiffness. This effect is detectable. Experimentally, it has been shown that when the laser intensity increase, the demagnetization time becomes longer [53, 54]. Koopmans et al [53] showed that increasing intensity slows down demagnetization in Co and Ni samples. The net change is 80 fs for both Ni and Co (Ni: from 140 to 220 fs; Co: 160 fs to 240 fs). However, the signal shape remains the same.

Our results suggest that this may be due to the exchange interaction reduction induced by the laser field instead of phonons, as shown by Lefkidis and Hübner [56]. This can be directly seen from equation (3), where a small exchange interaction leads to a weaker torque on the spin and prolongs the dynamics. Figure 7 shows an example in the ferromagnetic 40-site spin chain, where the demagnetization time $\Delta T$ at site 30 increases almost linearly with the exchange pulse amplitude $A$. Here the demagnetization time $\Delta T$ is defined as the difference between the first peak time of the $S_{8x}$ and the exchange pulse peak time $T$. The overall shape does not change strongly with the exchange pulse amplitude, similar to the experimental observation. Second, this effect must depend on the sample, since exchange interactions and magnetic spin moments are different in different samples. In Co/Pd multilayer films, Vodungbo et al [57] did not detect an obvious relation.

(see the thick solid line), with its period determined by the exchange interaction.

However, this is not the case for spin-frustrated orderings. For instance, configuration No. 805 has an in-plane antiferromagnetic ordering and an inter-plane ferromagnetic ordering. This structure is based on the AFM configuration and the only difference is that we introduce an inter-plane antiferromagnetic coupling. Without the exchange pulse, we find that the $x$-component of spin at site 8 shows a periodic oscillation from 0 fs to 600 fs (see the dotted line in figure 6(b)). If we turn on the exchange pulse at 200 fs, the overall change in $S_{8x}$ begins after 200 fs (see the solid line in figure 6(b)). There is no big change before 200 fs, even though the exchange pulse is already oscillating periodically with time, but with the exchange pulse, $S_{8x}$ changes. This is again opposite to the common belief that the spin dynamics due to the exchange interaction ceases to occur after the typical time of the exchange interaction and all the subsequent spin relaxation results from the phonon.

For other two configurations (Nos. 801 and 802), we reach the same conclusion. Figure 6(c) shows that without the exchange pulse, $S_{7y}$ shows a clear change, but when the exchange pulse is introduced, the change starts (see the solid line); the major change occurs around 350 fs, after which the exchange pulse is completely restored to $J_0$. In figure 6(d) we show the results for configuration No. 802. Without the exchange pulse, $S_{8x}$ already oscillates periodically with time, but with the exchange pulse, we see that the oscillation becomes aperiodic, with the extreme values of $-1h$ and $1h$. There is also a delay between the exchange pulse and the major spin change. This is the main finding of our present study.

4. Applications to experiments

4.1. Delay between the exchange pulse and spin dynamics

The delay between the exchange pulse and spin dynamics has important consequences for experiments. Theoretically, this delay is expected since the spin wave needs time to propagate from one site to the next. For a normal uniform magnetic ordering such as FM and AFM, the collective spin excitation can survive the exchange interaction change. Physically, they have a larger spin stiffness. This effect is detectable. Experimentally, it has been shown that when the laser intensity increase, the demagnetization time becomes longer [53, 54]. Koopmans et al [53] showed that increasing intensity slows down demagnetization in Co and Ni samples. The net change is 80 fs for both Ni and Co (Ni: from 140 to 220 fs; Co: 160 fs to 240 fs). However, the signal shape remains the same.

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between the pump fluence and the demagnetization time. This finding is very important, as it casts doubt on the phonon mechanism (see below for more).

For GdFeCo, the situation is more complicated and more interesting. Experimentally, Stanciu et al. [40] showed that even though their laser pulse is only 40 fs long, the spin switch occurs on a picosecond time scale. There is a long delay between the laser initial excitation and the actual spin switching. As seen from figure 6, the exchange pulse does not act on the spin immediately; there is a long delay before the actual spin starts to change. For GdFeCo, the exchange interaction is much weaker, on the order of 0.01 eV or smaller, so the delay can be much longer. Alebrand et al. [58] found that the helicity information stored in GdFeCo lasts at least some picoseconds after optical excitation. The information carrier must not be phononic, since lower temperature works better for switching as Hohlfeld showed [59]. This issue has been discussed extensively in our prior study [17].

Khorsand et al. [60] showed that the threshold fluence for optical switching is independent of the helicity of the laser pulse, but this result has not yet been reproduced by other groups. Theoretically, we find that in general the exchange interaction is reduced more with circular polarized light than with linearly polarized light, so that we expect it is easier to switch spins. With a strong laser, regardless of its helicity, an even larger reduction in exchange interaction is expected.

Experimentally, Ostler et al. [48] reported an intriguing result in GdFeCo that a minor increase in the laser intensity could change the spin switching mechanism from a helicity-dependent to helicity-independent switch. Before we compare with the experimental results, we would like to resolve some confusions in the literature. Second, in their supplementary material [48], we notice an important experimental detail [17]: increasing the laser intensity from 2.30 mJ cm$^{-2}$ to 2.25 mJ cm$^{-2}$ leads to a transition from a helicity-dependent to helicity-independent switching. Such a dramatic switch difference is interesting and puzzling.

Theoretically, we compute the spin moment change as a function of various exchange pulse amplitudes $A$ for both AFM (No. 806) and spin-frustrated orderings (No. 805). We change $A$ from 0.05 to 0.50. The exchange pulse has the same duration and delay as figure 6. Figure 8(a) shows the results for AFM ordering. We see that the effect on the spin change is very minor, only with the oscillation amplitude increased. However, for the spin-frustrated case (No. 806), we see a big difference. Figure 8(b) shows that when the exchange amplitude is small, the change is gradual, but when $A$ is above 0.40, every 0.05 increase in the exchange interaction leads to very different spin dynamics. Once $A$ is above 0.3, the spin switching from the negative maximum to the positive maximum occurs earlier for $A = 0.5$ than for $A = 0.3$, though both the spins reach the first negative maximum at the same time. While it is true that our model is still very different from the real experimental situation, qualitatively our result reveals the possibility to induce a dramatic change using a strong laser pulse. Additional study is needed to completely explain the experimental observations.

### 4.2. Phonon controversy

Many researchers strongly argue that slower demagnetization with a stronger laser is due to the phonon involvement [53, 62, 63] through the Elliot–Yafet (EY) effect [64, 65]. But Vodungbo et al. [57] failed to find any significant change in the demagnetization. Does this indicate no phonon involvement? Theoretically, we find this unsatisfactory. However, the main evidence against the EY mechanism is from the EY theory itself. Regardless of whether it is suitable for femtosecond or not, the theory predicts that the spin relaxation time $T_1$ is inversely proportional to the number of phonons through the equation,

$$T_1 \propto \int \int d\mathbf{k}d\mathbf{k}'G_{k,k'}|n_q\delta(E_{k\uparrow} - E_{k'\downarrow} + \hbar \omega_q) + (n_{-q} + 1)\delta(E_{k\downarrow} - E_{k'\uparrow} - \hbar \omega_q)|$$

where the integrations are over the crystal momentum index $k(k')$, $G_{k,k'}$ is the matrix element for the spin flip (see equation (18.4) of Yafet’s theory [65] for details), $n_q$ is the number of phonons with phonon momentum $q$, $\hbar \omega_q$ is the phonon energy and $E_{k\uparrow(\downarrow)}$ is the spin up(down) band energy. Since the number of the phonons increases with the laser intensity naturally, the demagnetization time would become shorter if the entire process were due to phonons. This is just opposite to what is observed experimentally.

To this end, it is clear that the EY mechanism contradicts the experimental results, but we wonder how much the spin moment is changed due to the lattice vibration. This has never
been done before. We take bcc Fe as an example. We construct a $2 \times 2 \times 2$ supercell (see the inset in figure 9), which has 16 Fe atoms in the cell. Our method is based on the linearized augmented plane-wave method as implemented in the WIEN2k code [45, 66]. We use the GGA functional and the $\mathbf{R} \mathbf{K}_{\text{max}}$ is 7.0. The total number of $\mathbf{k}$ points is 1000. We self-consistently solve the Kohn–Sham equation

$$[-\nabla^2 + V_{\text{Ne}} + V_{\text{ec}} + V_{\text{xc}}^i] \psi_{\mathbf{k}}^\sigma(r) = E_{\mathbf{k}}^i \psi_{\mathbf{k}}^\sigma(r), \quad (5)$$

where the terms on the left-hand side are the kinetic energy, electron-nuclear attraction, Coulomb and exchange interactions, respectively. $\psi_{\mathbf{k}}^\sigma(r)$ is the Bloch wavefunction of band $i$ at crystal momentum $\mathbf{k}$ with spin $\sigma$ and $E_{\mathbf{k}}^i$ is the band energy.

To compute the effect of the lattice vibration, we displace one Fe atom by 0.1 Å along the diagonal direction of the cube (see the inset again). The total energy change due to the Fe displacement is 0.481 eV, already over 10 times higher than the typical phonon energy. Figure 9 shows the atom-resolved spin moment change, with the same colors representing equivalent spin changes. The results are insightful. Only two atoms have a large spin moment change: the spin moment on No. 9 is reduced by $-0.08 \mu_B$, while that on No. 16 is increased by $0.05 \mu_B$. Both atoms are close to the displaced atom No. 1. Changes on other atoms are below 0.02 $\mu_B$. These changes are only 3.6%, in comparison to the pristine spin moment of 2.2 $\mu_B$. Therefore, a simple lattice vibration only leads to a very small spin moment change [67], which is not enough to explain the experimental findings.

### 5. Conclusion

We have proposed the laser-induced exchange interaction change as an alternative path to ferromagnetism. We show that in general the exchange interaction is reduced for the excited states and the reduction is helicity-dependent, with a larger reduction found with circularly polarized light than with linearly polarized light. Second, we introduce an exchange interaction pulse and investigate the spin dynamics in two systems with three magnetic ordering (FM, AFM and spin-frustrated). For pure FM and AFM, we find a clear delay of the spin dynamics with respect to the exchange pulse and the delay becomes longer with a stronger pulse, in agreement with the experimental results in 3d transition metals. A dramatic spin change is found in spin-frustrated systems and is also delayed by several hundred femtoseconds, which is consistent with the observation in GdFeCo. Finally, we examine the spin change with the exchange pulse amplitude. We find that for a weak pulse, the change is small. However, if the pulse is very strong such as $A = 0.4$, a change as small as 0.05 $\mu_B$ can potentially lead to a strong change. This nicely explains why experimentally [48] only a small increase in the laser intensity by 0.05 mJ cm$^{-2}$ could induce a helicity-independent switch. We believe that it is the exchange interaction reduction that plays a role here. We also explain why the phonon effect from the EY theory does not play a critical role here. Our first-principles calculation shows that even moving one iron atom by 0.1 Å along the lattice diagonal, the spin moment is only reduced by 0.1 $\mu_B$ out of 2.2 $\mu_B$. Our new mechanism is expected to inspire new theoretical [17, 19–21, 45, 46, 56, 62, 63, 67] and experimental investigations [18, 22, 24–27, 29, 32, 48, 58, 60, 61, 68].

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**Figure 8.** Intensity dependence of the spin switch. (a) For regular AFM ordering, the exchange pulse amplitude has no significant effect on the spin change. (b) For a frustrated-spin ordering (configuration number 805, see figure 5), when the amplitude is higher, even a small change in the exchange amplitude leads to a dramatic change in spin dynamics. This is consistent with the experimental observation. The numbers on each curve are the exchange pulse amplitude. All the curves are shifted vertically for easy viewing.

**Figure 9.** Effect of the atom vibration on the spin moment change in bcc Fe. A cell of $2 \times 2 \times 2$ is chosen. We move atom 1 (with arrow) by 0.1 Å along the diagonal direction and compute the spin moment change using the density functional theory. The largest spin moment change occurs for two atoms close to the first atom (see the red ($-0.08 \mu_B$) and dark green ($+0.05 \mu_B$) bars). It is obvious that the spin moment change is small.
state $\psi$ is well known, $x$ PIB. In this model, the potential is zero if the electron position compute the value of Coulomb and exchange interactions in To appreciate the change in electron-electron interactions, we in particle-in-a-box (PIB) Appendix B.. Electron–electron interaction change in particle-in-a-box (PIB).

To see why the optical transition imposes a restriction on the electron configuration, we compute the transition matrix elements $\langle \psi_{\text{exc}}(D) | \psi_{\text{gs}} \rangle$ for a transition from the ground state $\psi_{\text{gs}} = (\phi_a(1)\phi_b(2) - \phi_a(2)\phi_b(1))/\sqrt{2}$ to excited state $\psi_{\text{exc}} = (\phi_a(1)\phi_b(2) - \phi_a(2)\phi_b(1))/\sqrt{2}$. A straightforward calculation yields

$$
\langle \psi_{\text{exc}} | D | \psi_{\text{gs}} \rangle = \langle \phi_a | D | \phi_b \rangle \delta_{ab} + \langle \phi_a | D | \phi_b \rangle \delta_{ac} - \langle \phi_a | D | \phi_b \rangle \delta_{ad} - \langle \phi_a | D | \phi_b \rangle \delta_{db}.
$$

(A.1)

These delta functions restrict the transition to occur only with one single-particle wavefunction changed. As a result, the exchange integral in the excited state has only four independent terms: $-\langle \phi_a \phi_b | r_{12}^{-1} | \phi_b \phi_a \rangle$, $-\langle \phi_a \phi_b | r_{12}^{-1} | \phi_b \phi_a \rangle$, $-\langle \phi_0 \phi_d | r_{12}^{+1} | \phi_d \phi_0 \rangle$ and $-\langle \phi_0 \phi_d | r_{12}^{+1} | \phi_d \phi_0 \rangle$. All the other ones are forbidden.

Appendix B.. Electron–electron interaction change in particle-in-a-box (PIB)

To appreciate the change in electron-electron interactions, we compute the value of Coulomb and exchange interactions in PIB. In this model, the potential is zero if the electron position $x$ is within $[0, L]$ and is infinite if otherwise. The eigenfunction is well known,

$$
\phi_n = \sqrt{\frac{2}{L}} \sin \left( \frac{n \pi x}{L} \right),
$$

(B.1)

where $n$ is the state index. To overcome the singularity in the Coulomb potential, we introduce a small broadening $\delta = 0.01$ (in the unit of $L$). This broadening affects the absolute value, but not the qualitative change of our results, which is verified by using different values of $\delta$. Our integrals have the following form:

$$
U(a|b|c|d) = \int_0^L \int_0^L dx_1 dx_2 \phi_n^*(x_1) \phi_n^*(x_2) \times \frac{e^2}{|x_1 - x_2| + \delta} \phi_0(x_2) \phi_0(x_1).
$$

(B.2)

References

[1] Stöhr J and Siegmann H C 2006 Magnetism: from Fundamentals to Nanoscale Dynamics (Berlin: Springer)
[2] Slater J C 1936 Phys. Rev. 49 537
[3] Brooks H 1940 Phys. Rev. 58 909
[4] Herring C and Kittel C 1951 Phys. Rev. 81 869
[5] Kaplan H 1952 Phys. Rev. 85 1038
[6] Wohlfarth E P 1953 Rev. Mod. Phys. 25 211
