An attempt to simulate laser-induced all-optical spin switching in a crystalline ferrimagnet

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Interest in all-optical spin switching (AOS) is growing rapidly. The recent discovery of AOS in Mn$_2$RuGa provides a much needed clean case of crystalline ferrimagnets for theoretical simulations. Here, we attempt to simulate it using the state-of-the-art first-principles method combined with the Heisenberg exchange model. We first compute the spin moments at two inequivalent manganese sites and then feed them into our model Hamiltonian. We employ an ultrafast laser pulse to switch the spins. We find that there is a similar optimal laser field amplitude to switch spins. However, we find that the exchange interaction has a significant effect on the system switchability. Weakening the exchange interaction could make the system unswitchable. This provides a crucial insight into the switching mechanism in ferrimagnets.

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Central to the magnetic storage device is the writing/reading speed of magnetic bits in a storage medium. Traditionally, these operations are mostly driven by an external magnetic field. A full-optical driven spin manipulation could break the speed barrier of several hundred picoseconds set by the Zeeman interaction and magnetic dipole-dipole interaction. In 1996, Beaurepaire et al. [1] showed that when they shone a 60-fs pulse on the ferromagnetic nickel thin film, they found a sharp decrease in the Kerr signal within 1 ps. This finding received immediate attention worldwide, and a new research field, femtosecond magnetism, was born [2, 3]. Research intensified, and is far beyond the scope of the original research interest. In 2007, Stanciu et al. [4] showed that a left-circularly polarized laser pulse can switch a down-spin to up, while a right-circularly polarized laser pulse can switch a down-spin up. This remarkable property represents an interesting new magnetic phenomenon on an ultrafast time scale, although their compound, GdFeCo, is not new. GdFeCo has been used in traditional magneto-optical recording (see the references cited in [2, 3]). However, being able to switch spins on a picosecond time scale optically is new, and has raised the possibility for a real application. However, it is unclear how the laser pulse can switch spins directly. Ostler et al. [5] further showed that if the laser intensity is increased above a certain level, regardless of laser helicity, each pulse can flip spins from one direction to another deterministically. They argued that there is a threshold intensity that one has to exceed to change from all-optical helicity dependent spin switching to all-optical helicity independent switching, but the actual picture is more complicated [6, 7].

For a long time, GdFeCo was the only material that shows AOS. Soon, many more materials were found [8–14]. However, these materials are mostly amorphous, which introduces an uncertainty in theoretical simulations and represents a formidable task. In 2017, Vomir et al. [15] reported the first observation of AOS in a Pt/Co/Pt ferromagnetic stack, but the switching is not complete. Very recently, Banerjee et al. [16] showed single-pulse all-optical toggle switching of magnetization in Mn$_2$RuGa. Mn$_2$RuGa is a ferrimagnetic Heusler compound, with a cubic structure. Two Mn atoms are not equivalent, and have different spin moments. They are antiferromagnetically coupled. As shown before [17], ferrimagnets have a big advantage over ferromagnets and antiferromagnets. This offers an ideal theoretical model.

In this paper, we investigate all-optical switching in Mn$_2$RuGa. Different from prior studies, we compute the spin moments at two Mn sites using the first-principles density functional theory. These spin moments are fed into the Heisenberg exchange model with both spin-orbit coupling and a harmonic potential [18, 19]. We find that not any arbitrary laser field amplitude can switch spins. There is a narrow window of opportunity where the spins at two Mn sites can be switched into their respective opposite directions. Because of the strong spin moments at two Mn sites, its switching is very stable. Quite different from other systems, we find that if we reduce the exchange interaction, the spins precess strongly at both Mn sites, very much like a regular antiferromagnet, in-
stead of a ferrimagnet. These strong spin oscillation occur at both Mn$_1$ and Mn$_2$ sites. Their oscillation period is inversely proportional to the laser field amplitude, similar to the Rabi frequency in a two-level system. For a weak exchange interaction case, regardless of the magnitude of the laser field amplitude, the spin switching is not observed. This points out an entirely different scenario from ferromagnetic cases \cite{14} and demagnetization \cite{20}. The rich picture that is found here reveals a crucial effect of exchange interaction on AOS, and should motivate further experimental and theoretical investigations in the future.

Mn$_2$RuGa is a Heusler ferrimagnet, with a stoichiometric composition of $X_2YZ$ and space group $F43m$. Two Mn atoms, Mn$_1$ and Mn$_2$, are situated at (4a) and (4c), which are magnetically inequivalent \cite{21 22}. Their spins are antiferromagnetically coupled. To properly investigate magnetic properties of Mn$_2$RuGa, we employ the density functional theory using the full-potential augmented plane wave method as implemented in the Wien2k code \cite{23 24}. Our first-principles calculation shows that Mn$_1$ has a spin moment of 3.17232 $\mu_B$ and Mn$_2$ has -2.30765 $\mu_B$. This agrees with prior calculations \cite{21 22}. So each cell has a net spin moment of 1.02394 $\mu_B$, close to unity, which is consistent with the nature of a stoichiometric half-metal \cite{25}. The spin moments on Ru and Ga are very small, and will be ignored below.

It is not often recognized that the large spin moments on Mn atoms are advantageous since the spin-orbit torque is proportional to the spin moment \cite{18 19}. Since it is not possible to simulate all-optical spin reversal at the first-principles level, in the following we will feed these two spin moments into our Heisenberg-exchange coupled harmonic model \cite{17 19 26 27}, and limit ourselves to a small system with 101 lattice sites along the $x$ axis and $y$ axis, respectively, with two monolayers along the $z$ axis. Our Hamiltonian is

$$H = \sum_i \left[ \frac{p_i^2}{2m} + V(\mathbf{r}_i) + \lambda \mathbf{L}_i \cdot \mathbf{S}_i - e\mathbf{E}(\mathbf{r}, t) \cdot \mathbf{r}_i \right] - \sum_{ij} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j,$$

(1)

where terms from the left to right are respectively the kinetic energy operator of the electron, the potential energy operator, the spin-orbit coupling, the interaction between the laser and system, and the exchange interaction between spins. Our exchange parameter $J_{ij}$ is still time-independent, although prior studies have shown that the exchange interaction itself could be affected by the electric field \cite{28 29}. $\lambda$ is the spin-orbit coupling constant, $\mathbf{L}_i$ and $\mathbf{S}_i$ are the orbital and spin angular momenta at site $i$, respectively, and $\mathbf{p}$ and $\mathbf{r}$ are the momentum and position operators of the electron, respectively. We choose a spherical harmonic potential $V(\mathbf{r}_i) = \frac{1}{2}m\Omega^2 r_i^2$ with system frequency $\Omega$. $\mathbf{E}(\mathbf{r}, t)$ is the laser field. This model is the only magnetic field-free model currently available to simulate spin reversal, while the commonly used model employs an effective magnetic field \cite{3}, which should be avoided. It represents a small step towards a complete model.

In order to compute the spin change, we solve the Heisenberg equation of motion \cite{26} for each spin operator at every site under laser excitation \cite{17}. Figure \(1\) shows the spin $z$ component at two Mn sites as a function of time. We employ a laser pulse of 60 fs, with a field amplitude of 0.017 V/A. We see that the spin at the Mn$_1$ site starts from the positive $z$ axis (see the circles). Upon laser excitation, it switches over the negative $z$ axis, while the spin at the Mn$_2$ site switches up from its $-z$ direction. This is consistent with the experimental observation \cite{15}. The strong spin moment stabilizes the entire switching process. However, not any arbitrary laser field amplitude can lead to faithful switching. Figure \(1\) shows how the final spin changes with the laser field amplitude. The dependence is highly nonlinear. If we use a weak laser pulse, there is little change in spins at both sites. But if the laser field is too strong, the spins overturn toward the $xy$ plane, so there is no spin reversal either. We find that the optimal field amplitude is 0.017 V/A, whose result is shown in Fig. \(1\). In this regard, Mn$_2$RuGa is pretty much similar to other ferrimagnets where there is an optimal amplitude \cite{17 19}. Microscopically, the real situation is more complicated.

To this end, there is no generic understanding of spin switching in both ferromagnets \cite{14 15} and ferrimagnets \cite{4}. It has been often argued that the angular momentum exchange between two spin sublattices in ferrimagnetic GdFeCo \cite{30} is the key to AOS. Theoretically, this momentum exchange picture is interesting, but such momentum exchange between sublattices, if it exists, occurs all the time through the exchange interaction, with or without the laser. In other words, it must be something extra due to the laser that switches the spin. In GdFeCo, a rigorous testing is difficult because it is amorphous, and it is difficult to tell whether a model system really represents a true GdFeCo sample. This brings ambiguity to a theoretical simulation. Mn$_2$RuGa removes this ambiguity completely.

As a first test, we investigate the effect of the exchange interaction on AOS. We reduce the exchange interaction from 0.1 eV to 0.001 eV. From prior studies, we know such a reduction does not constitute a major issue for
A creases with laser field amplitude of the spin, respectively. (b) The spin oscillation period changes on spin reversal is much more pronounced and minimum spin values, respectively. The effect of the exchange interaction on spin reversal is not found. Only a strong field amplitudes, spin reversal is not found. Only a strong exchange interaction. Here we choose a narrow window that one can switch spins.

FIG. 1: (a) The z component of the spins at the Mn_1 and Mn_2 sites as a function of time at the optimal laser field amplitude. Here, the laser amplitude is 0.017 V/Å, and the pulse duration is 60 fs. The empty circles denote the spin at the Mn_1 site, while the boxes refer to the spin at the Mn_2 site. We see there is a clear spin reversal upon laser excitation. (b) Dependence of the S_z as a function of the laser field amplitude A_0. The empty circles refer to the spin at Mn_1 site, and the boxes refer to the spin at Mn_2 site. A weak laser field does not reverse the spins, but a too strong laser field can not either. There is a narrow window that one can switch spins.

FIG. 2: (a) Spin precession at Mn_1 site under a reduced exchange interaction. Here we choose J = 0.001 eV. The rest of parameters are the same as those in Fig. 1. The solid, dotted, and dashed lines denote the x, y, and z components of the spin, respectively. (b) The spin oscillation period decreases with laser field amplitude A_0. (c) At any of the laser field amplitudes, spin reversal is not found. Only a strong oscillation is noticed. The solid line is the time-average of the spin, and the dotted and dashed lines refer to the maximum and minimum spin values, respectively. The effect of the exchange interaction on spin reversal is much more pronounced in Mn_2RuGa than in other materials.

demagnetization in a system with a small spin moment. Figure 2(a) shows the spin change at Mn_1 (the situation is similar at Mn_2), where the solid, dotted, and dashed lines denote the x, y, and z components, respectively. Both the laser duration and amplitude are exactly the same as those in Fig. 1. We see that there is a strong oscillation in all these three components. We note in passing that these three components must obey the operator permutation [8], where they can not be considered a linear reversal [31]. In principle, we need to cut off the simulation around 1 ps, after which we need to introduce damping, but to demonstrate the high accuracy of our calculation, we do not use the damping. These strong oscillations resemble a pure antiferromagnetic case. The spins at two neighboring sites are out of phase and remain antiferromagnetically coupled, even upon laser excitation. The laser pulse essentially initiates the spin dynamics, and the exchange interaction takes over, without switching the spins. For this reason, the angular momentum exchange picture for AOS can not explain this even in the same ferrimagnet. The period of the oscillation is not determined by the exchange interaction and spin moment alone. Figure 2(b) shows that as we increase the laser field amplitude, the period becomes shorter. The small fluctuation at the largest amplitudes is due to the period sampling because the oscillation is not strictly harmonic. This laser-field dependence of the oscillation period is very similar to the Rabi period dependence. For all the field amplitudes that we investigate, we do not see a case where the spins are reversed. Figure 2(c) illustrates the average (solid line), maximum (dotted line), and minimum (dashed line) of the final spin. We see the average spin never becomes negative (the initial spin is along the +z axis). The maximum and minimum values show the limits of spin. Our results point out an important fact: In a ferrimagnet, the effect of the exchange interaction is far more complicated than thought.

Now, we have two cases: One shows AOS, and the other does not. We can directly check whether the prior criteria proposed by Mentink et al. [8] apply to them. Their argument is based on a two-spin system, so for the pure exchange interaction, the spins at two sublattices must obey the scalar form of spins, \( \partial S_1/\partial t = -\partial S_2/\partial t \), with the extra term from demagnetization. In our system, each spin is coupled with more than four neighboring spins, so we take two neighboring spins as an example. For the above nonswitchable case (Fig. 2), we find that \( S_{1x} + S_{2x} \) and \( S_{1y} + S_{2y} \) are not constant, so they do not obey \( \partial S_1/\partial t = -\partial S_2/\partial t \). Our \( S_{1x} + S_{2x} \) decreases from 0 to about \(-1\) with oscillations, while \( S_{1y} + S_{2y} \) increases from 0 to about \(+1\) at the same rate. For our switchable case (Fig. 1), \( \partial S_1/\partial t = -\partial S_2/\partial t \) is not fulfilled either. Instead, we find that our result obeys the vector form \( S_1(t) = -S_2(t) \). This shows that the simple argument based on a two-spin model is not applicable to our realistic case. We plan to investigate this issue further in a much larger system.

In conclusion, we have carried out a joint first-
principles density functional theory and model simulation of all-optical spin reversal in Mn$_2$RuGa. We are able to find a case that the spins can be switched without employing a magnetic field. The system also shows an optimal laser electric field amplitude, with the same profile like those in other systems. The spins at Mn$_1$ site are switched from the $+z$ axis to $-z$ axis, while those at Mn$_2$ site are switched from the $-z$ axis to $+z$ axis. This is fully consistent with the experimental findings. We find that the exchange interaction has a significant effect on the switching. When we reduce the exchange to 0.001 eV, we find the system becomes unswitchable. It behaves like a regular antiferromagnet. Our finding is expected to motivate further theoretical and experimental investigations in the future.

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Availability of data. The data that support the findings of this study are available from the corresponding author upon reasonable request.

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