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Peer Review File

Polarisation-dependent single-pulse ultrafast optical switching of an elementary ferromagnet

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

All-optical switching of magnetization provides a route to achieve ultrafast and energy-efficient magnetic storage. Though all-optical switching of magnetization in ferromagnetic materials has been widely studied, the underlying mechanism is still under debate. This manuscript reports a realistic time-dependent electronic structure simulation of laser-induced ultrafast magnetization dynamics in bulk Ni. The authors show sub-picosecond switching of magnetization by a single laser pulse, and detailed underlying physics is discussed. Although it cannot directly explain the current experimental observation in ferromagnets, it provides some new perspectives into this field. Before I make any recommendation, I would like the authors to address some critical questions and comments.

1. It looks that all-optical switching happens at a short timescale less than 6 ps, as shown in Figure 2d. However, at a longer timescale, the magnetization returns to the initial direction of the moment, as shown in Figure 3c and 3d. What is the reason, and does this phenomenon always happen for all cases at a longer timescale? If this is the case, the magnetization is not switched in the end.

2. On page 8, it is stated that "…the probability of spin reversal is high". Does the possibility come from several trials of calculation? If so, are the switching curves in Figure 2d always same or not?

3. On page 11, the authors provide several mechanisms for laser-induced magnetization dynamics. For circularly polarized light illumination, the optical spin-orientation and related optical spin transfer torque are also possible. It would be helpful that the authors can elaborate the discussions with some references.

4. As the authors stated, the time-dependent magnetic anisotropy induced torque acts after the laser-pulse is over, and it is related to the nonequilibrium electronic system. Since dissipation channel is not included in the modeling, the nonequilibrium will stay. It will determine the time and strength of MAE related torque, and overall magnetic dynamics in the end. It is possible that you cannot get switching if such torque is shorted and dampened.

5. I recommend the authors to add some recent papers on the related topic, including "Ab Initio Study of Helicity-Dependent Light-Induced Demagnetization: From the Optical Regime to the Extreme Ultraviolet Regime", Nano Lett. 21, 1943 (2021); "Inertial spin dynamics in ferromagnets", Nature Phys. 17, 245 (2021); "Ultrafast optical manipulation of magnetic order in ferromagnetic materials", Nano Convergences 7, 1 (2021); "All-Optical Manipulation of Magnetization in Ferromagnetic Thin Films Enhanced by Plasmonic Resonances", Nano Letters, 20, 6437 (2020); "All-Optical Helicity-Dependent Switching in Hybrid Metal–Ferromagnet Thin Films" Adv. Opt. Mater. 8, 2000379 (2020); "Helicity-Preserving Metasurfaces for Magneto-Optical Enhancement in Ferromagnetic [Pt/Co]N Films" Adv. Opt. Mater. 18, 2001420.

Reviewer #2 (Remarks to the Author):

The authors present a theoretical paper claiming that all-optical switching, using a single circularly-polarized pulse, can be achieved in an elementary ferromagnet such as Ni. They present a detailed phase diagram for pulse-duration vs absorbed fluence, and show that switching is achieved using a good combination of these two parameters. The calculations are performed using time-dependent electronic structure simulations, and reveal that the switching arises by laser-driven torques (from the circular polarization) that accompany the ultrafast demagnetization.

The field of all-optical switching is popular in the research community of ultrafast magnetism, and many works have been devoted to understanding the single-shot process in GdFeCo alloys. Experimental and theoretical works both emphasize that the switching there is rooted in the antiferromagnetic coupling between the Gd and FeCo sublattices. A similar explanation can be used for the same switching process recently discovered in MRG [Ref 12]. The pulse-duration has been shown to be crucially important in achieving this switching.

In ferromagnets, the lack of antiferromagnet coupling means that the switching process must be different in its physical origin. As the authors correctly point out, switching in the ferromagnet Co/Pt requires multiple circularly-polarized pulses. The most recent experimental work on Co/Pt [Ref 20] suggests that the switching can be achieved using two pulses. The first pulse basically acts as an ultrafast heater, demagnetizing the sample, and the second circularly-polarized delivers a torque that rotates the almost-quenched magnetization towards its switched orientation. It is not possible to achieve these effects simultaneously using one pulse since the demagnetization requires an ultrafast (fs) pulse, but the laser-induced torque demands a longer stimulus (several picoseconds) to increase its strength.

The discovery by the authors that single-shot switching could be achievable in a ferromagnet is therefore timely and will be of interest to experimentalists. However, unfortunately I do not think that the article (as it stands) deserves to be published in Nature Communications Physics. The problem is that this is an entirely-theoretical work that would be rather easy to experimentally prove or disprove. To test the prediction of all-optical switching in e.g. fcc Ni, one only needs to 1) find magneto-optical contrast using the Kerr effect in an imaging geometry, and 2) illuminate the material with a single circularly-polarized pulse of certain duration / fluence, as specified in Fig. 1b. Such experiments are routinely done by a large number of research groups around the world, including the groups of Jeff Bokor (Berkeley), Theo Rasing (Nijmegen), Bert Koopmans (Eindhoven), Martin Aeschlimann (Kaiserslautern), Stéphane Mangin (Nancy) etc. This experiment requires only a simple magneto-optical imaging setup and an amplified laser source, which is even available at Julich (http://dx.doi.org/10.17815/jlsrf-6-174).

The practical discovery of single-pulse all-optical switching in an elementary ferromagnet would represent a breakthrough that is probably worthy of publication in Nature Comms or even a journal with even higher impact (e.g. Nature Physics). I would therefore recommend the authors to attempt the experiment – without such experimental proof, the manuscript can only be published in a more specialized journal such as Phys Rev B or J. Phys. D: Cond. Matter. If the authors can indeed prove that their calculations are grounded in reality, the manuscript (including the experimental proof) should be submitted to e.g. Nature Communications.

Beyond my overall assessment, I have several comments and questions:

1. Line 38: The three-temperature model describes the demagnetization of bulk Ni, but cannot explain on its own the switching in GdFeCo. To achieve the latter, one needs a four-temperature model (taking in to account the inter-sublattice coupling between the spins of Gd and FeCo), as shown by e.g. A. Mekonnen et al, PRB 87, 180406 (2013).

2. Despite many attempts, the process of single-shot switching in ferromagnets (e.g. Co/Pt, FePt) has not been experimentally discovered. We always need to use several pulses. Can the authors therefore explain why the mechanism is different in Ni? Am I correct in understanding that the key difference lies in the in-plane orientation of the anisotropy? The authors should highlight this more clearly in the introduction.

3. The authors use a photon energy of 1.55 eV in their calculations. Is this an important parameter? Recent work has shown that the photon energy is not very important in the single-shot switching process in MRG [Ref 12], so does the photon energy play an important role here?

Reviewer #3 (Remarks to the Author):

The authors of the manuscript "Polarisation-dependent single-pulse ultrafast optical switching of an elementary ferromagnet" use a time-dependent tight-binding implementation to study the real-time dynamics of torque and magnetization in bulk nickel, following laser excitations of different polarization, pulse width, and fluence. From their data they conclude that the magnetization in the material can be switched by a single laser pulse and they report the parameter range for the laser excitation. In addition, by decomposing the results with respect to atom and angular momentum component the authors obtain insight into the underlying mechanism. I find this work interesting and important and I recommend its publication in Communications Physics. Prior to publication, I recommend clarification of the following points:

I highly suggest that that authors comment a bit more on error bars/limitations of their framework. For instance, to what extent does their approach model excitations of the magnetic subsystem? Or are these purely electronic excitations/transitions that are shown and cause the observed behavior? Does this framework allow for fully non-collinear magnetic configurations based on Dirac four vectors, especially during switching?

In addition, the manuscript, at times, strongly focuses on describing figures, but it is not so clear what physics can be learned or what the underlying takehome message is, e.g. around line 161 on p 11. On line 169, p. 13, the authors mention that the time scale is connected to magnetic anisotropy energy, but do not elaborate. Regarding time scales, the comments on lines 203, 212, and 216 can also be made more specific/shown in more detail. On line 192, p. 15, the authors discuss that the coupling is "well known", but do not provide a reference. Similarly, wording/clarity can be improved in many places.

Finally, to strengthen their conclusions, the authors discuss an interesting mechanism around line 199, and it would be very helpful if they could elaborate on its general characteristics and importance. How are the results obtained in this work transferable to other material systems?

In addition, there are the following minor points:

The authors refer to as "specific algorithm enabling to monitor the non-linear magnetisation dynamics", but seemingly without details?

The authors also claim that their approach enables them "to use larger pulse widths". Details are needed. Larger relative to what?

On line 123 of page 9, the authors mention a minimum switching threshold that is reasonable and expected, but they do not elaborate on why this is expected and what exactly is expected.

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In the following we reply point by point to the matters raised by both reviewers. The original reports are shown in black, and our replies in blue. The modified text from the manuscript is shown in red.

Reviewer 1 (Comments to the Author):

All-optical switching of magnetization provides a route to achieve ultrafast and energy-efficient magnetic storage. Though all-optical switching of magnetization in ferromagnetic materials has been widely studied, the underlying mechanism is still under debate. This manuscript reports a realistic time-dependent electronic structure simulation of laser-induced ultrafast magnetization dynamics in bulk Ni. The authors show sub-picosecond switching of magnetization by a single laser pulse, and detailed underlying physics is discussed. Although it cannot directly explain the current experimental observation in ferromagnets, it provides some new perspectives into this field. Before I make any recommendation, I would like the authors to address some critical questions and comments.
We thank the reviewer for identifying the relevance and purpose of our study, and for supporting its publication after her/his questions and comments.

1. It looks that all-optical switching happens at a short timescale less than 6 ps, as shown in Figure 2d. However, at a longer timescale, the magnetization returns to the initial direction of the moment, as shown in Figure 3c and 3d. What is the reason, and does this phenomenon always happen for all cases at a longer timescale? If this is the case, the magnetization is not switched in the end.
The reviewer is right to point out that in some cases the magnetization returns to the hemisphere with $z > 0$ (but not necessarily the same direction) after some time. There are however cases, where up to the limit of the simulation time, the magnetization oscillates after crossing the zero line (so not around zero), and keeps pointing at negative z. Owing to the absence of dissipation mechanisms in our calculations, the energy introduced to the system by the laser field can keep the magnetization oscillating for longer time scales. Our reasoning is that after a few picoseconds the dissipation [Phys. Rev. Lett. 107, 207201 (2011), Science Advances 5, eaau8000 (2019), Phys. Rev. B 101, 100302(R) (2020)] will prevent these long time oscillations, and the longer the magnetization stays in the hemisphere with $z < 0$, the higher the probability for the system to relax to the switched direction.

2. On page 8, it is stated that "…the probability of spin reversal is high". Does the possibility come from several trials of calculation? If so, are the switching curves in Figure 2d always same or not?
The calculations are deterministic: for a given set of parameters, the obtained switching curves are the same. By "probability" we refer to the oscillations that the reviewer pointed out in the previous question: the green region indicates the range of parameters for which the system switches direction, but could return to the original one after some time. We have added the following to the manuscript:
We note that the magnetization in some cases can switch back after a few ps and then experiences an oscillatory behavior. This is the time scale where the dissipation mechanisms, not included in our simulations, kick in, which we believe will help stabilizing the magnetization switching.

3. On page 11, the authors provide several mechanisms for laser-induced magnetization dynamics. For circularly polarized light illumination, the optical spin-orientation and related optical spin transfer torque are also possible. It would be helpful that the authors can elaborate the discussions with some references.
The reviewer rightly points out that the circular polarisation of the laser enables the optical spin transfer torque.

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In fact, our simulations show that the circular polarisation is a key ingredient to achieve an efficient switching of the magnetisation direction. We write in the Introduction: "The inverse Faraday effect was proposed as a direct mechanism for laser-induced demagnetisation [Ref. 29], which evolved into a more general picture of light-induced magnetic torques [Refs. 30–35]." Those references also cover the optical spin transfer torque. It is this more general picture of light-induced magnetic torques that we invoke in Eq. 1 and in Fig. 4a, and we use "inverse Faraday effect" as an umbrella term to convey this meaning.

4. As the authors stated, the time-dependent magnetic anisotropy induced torque acts after the laser-pulse is over, and it is related to the nonequilibrium electronic system. Since dissipation channel is not included in the modeling, the nonequilibrium will stay. It will determine the time and strength of MAE related torque, and overall magnetic dynamics in the end. It is possible that you cannot get switching if such torque is shorted and dampened.
The scenario described by the reviewer is indeed possible and is for sure material dependent. However, as we argued in the reply to the first question, we expect that the timescale associated with such damping-like torques will still enable the initial switching of the magnetization direction, with the added benefit of suppressing subsequent oscillations of the magnetization towards its initial orientation.
For instance, the role of phonons as a channel for angular momentum dissipation has been very much debated. For instance, Oppeneer and coworkers [Phys. Rev. Lett. 107, 207201 (2011)] theoretically showed that Elliot-Yafet phonon scattering does not enter the dynamics at an early stage, while the ab-initio simulations of Chen and Wang [Science Advances 5, eaau8000 (2019)] demonstrated that phonons is not the dominant effect in the demagnetization although it is active within a few hundred fs. On the experimental side, Maldonado et al. [Phys. Rev. B 101, 100302(R) (2020)] recently showed that phonons are active within a few ps.
We added the references above in the final discussion of our manuscript.

5. I recommend the authors to add some recent papers on the related topic, including - "Ab Initio Study of Helicity-Dependent Light-Induced Demagnetization: From the Optical Regime to the Extreme Ultraviolet Regime", Nano Lett. 21, 1943 (2021); "Inertial spin dynamics in ferromagnets", Nature Phys. 17, 245 (2021); "Ultrafast optical manipulation of magnetic order in ferromagnetic materials", Nano Convergences 7, 1 (2021); "All-Optical Manipulation of Magnetization in Ferromagnetic Thin Films Enhanced by Plasmonic Resonances", Nano Letters, 20, 6437 (2020); "All-Optical Helicity-Dependent Switching in Hybrid Metal-Ferromagnet Thin Films" Adv. Opt. Mater. 8, 2000379 (2020); "Helicity-Preserving Metasurfaces for Magneto-Optical Enhancement in Ferromagnetic [Pt/Co]N Films" Adv. Opt. Mater. 18, 2001420.
We thank the reviewer to point us to those references. We have included them in our manuscript.

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Reviewer 2 (Comments to the Author):

The authors present a theoretical paper claiming that all-optical switching, using a single circularly-polarized pulse, can be achieved in an elementary ferromagnet such as Ni. They present a detailed phase diagram for pulse-duration vs absorbed fluence, and show that switching is achieved using a good combination of these two parameters. The calculations are performed using time-dependent electronic structure simulations, and reveal that the switching arises by laser-driven torques (from the circular polarization) that accompany the ultrafast demagnetization.
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In ferromagnets, the lack of antiferromagnet coupling means that the switching process must be different in its physical origin. As the authors correctly point out, switching in the ferromagnet Co/Pt requires multiple circularly-polarized pulses. The most recent experimental work on Co/Pt [Ref 20] suggests that the switching can be achieved using two pulses. The first pulse basically acts as an ultrafast heater, demagnetizing the sample, and the second circularly-polarized delivers a torque that rotates the almost-quenched magnetization towards its switched orientation. It is not possible to achieve these effects simultaneously using one pulse since the demagnetization requires an ultrafast (fs) pulse, but the laser-induced torque demands a longer stimulus (several picoseconds) to increase its strength.
The discovery by the authors that single-shot switching could be achievable in a ferromagnet is therefore timely and will be of interest to experimentalists. However, unfortunately I do not think that the article (as it stands) deserves to be published in Nature Communications Physics. The problem is that this is an entirely-theoretical work that would be rather easy to experimentally prove or disprove. To test the prediction of all-optical switching in e.g. fcc Ni, one only needs to 1) find magneto-optical contrast using the Kerr effect in an imaging geometry, and 2) illuminate the material with a single circularly-polarized pulse of certain duration / fluence, as specified in Fig. 1b. Such experiments are routinely done by a large number of research groups around the world, including the groups of Jeff Bokor (Berkeley), Theo Rasing (Nijmegen), Bert Koopmans (Eindhoven), Martin Aeschlimann (Kaiserslautern), Stéphane Mangin (Nancy) etc. This experiment requires only a simple magneto-optical imaging setup and an amplified laser source, which is even available at Julich (http://dx.doi.org/10.17815/jlsrf-6-174).
We thank the reviewer for carefully reading our manuscript and for sharing our enthusiasm concerning the importance of our results. Our work is purely theoretical, obtained with state of the art time-dependent electronic structure simulations based on a computational code that we recently developed for this very purpose. As the reviewer highlights, the implications of our predictions will certainly trigger exciting experiments from various experimental groups leading the field of ultrafast magnetization dynamics and will resonate within this highly competitive field of research. Owing to the timeliness and importance of our results, we firmly believe that our theoretical work deserves publication in Communications Physics. We are actively disseminating our results at various conferences and workshops and anticipate future collaborations with leading experimental groups.

The practical discovery of single-pulse all-optical switching in an elementary ferromagnet would represent a breakthrough that is probably worthy of publication in Nature Comms or even a journal with even higher impact (e.g. Nature Physics). I would therefore recommend the authors to attempt the experiment – without such experimental proof, the manuscript can only be published in a more specialized journal such as Phys

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Rev B or J. Phys. D: Cond. Matter. If the authors can indeed prove that their calculations are grounded in reality, the manuscript (including the experimental proof) should be submitted to e.g. Nature Communications.
We appreciate very much that the reviewer finds our predictions to be a breakthrough in the field and the recommendation to publish our work in prestigious journals. We believe that Communications Physics is a nice avenue for our theoretical manuscript. In the future, we plan in extending this work to involve experimental measurements from various groups, but this goes beyond the manuscript currently under review.

Beyond my overall assessment, I have several comments and questions:
1. Line 38: The three-temperature model describes the demagnetization of bulk Ni, but cannot explain on its own the switching in GdFeCo. To achieve the latter, one needs a four-temperature model (taking in to account the inter-sublattice coupling between the spins of Gd and FeCo), as shown by e.g. A. Mekonnen et al, PRB 87, 180406 (2013).
We thank the reviewer for pointing out our incorrect referencing. We have modified the text and reference to: "The three-temperature model describes the nonequilibrium thermodynamics of coupled electronic, magnetic and lattice subsystems, providing a very good semi-phenomenological description of the demagnetisation of bulk Ni. It can also be generalized to describe the switching of ferrimagnetic GdFeCo [A. Mekonnen et al, PRB 87, 180406 (2013)].".

2. Despite many attempts, the process of single-shot switching in ferromagnets (e.g. Co/Pt, FePt) has not been experimentally discovered. We always need to use several pulses. Can the authors therefore explain why the mechanism is different in Ni? Am I correct in understanding that the key difference lies in the in-plane orientation of the anisotropy? The authors should highlight this more clearly in the introduction.
We thank the reviewer for the very interesting question. We are currently studying Co/Pt, and Pt plays a crucial role in the dynamical behavior of such a material. A definite reason cannot be given right now without a careful investigation, which we are undertaking. What we can say is that the magnetic anisotropy energy is much weaker in Ni than in Co/Pt or FePt, and we remark that it has cubic symmetry for the former and uniaxial symmetry for the latter two. For bulk Ni, we believe that an important aspect, which is discussed in detail in Fig. 5 of our manuscript, is the antiferromagnetic coupling between the different orbitals of Ni, which gets altered after the material is excited by the electric field through two mechanisms. The first is the intra-orbital spin-flips on the time scale of the spin-orbit coupling which causes the initial reduction of the magnetization carried by the sp-electrons. The second mechanism is due to the alteration of the exchange coupling strength between the sp and d-electrons due to the orbital repopulation, which drives the magnetization of the d-orbitals to grow in a direction opposite to that of sp-orbitals, and so, the antiferromagnetic alignment is restored by the end of the pulse duration. This leads to a rich orbital-dependent transient dynamics involving intra-atomic non-collinear magnetic states. Based on our preliminary simulations on Co/Pt, other mechanisms come into play such as ultrafast Hall effects in conjunction with the inverse-Faraday effect, which dictate complex magnetization dynamics.

3. The authors use a photon energy of 1.55 eV in their calculations. Is this an important parameter? Recent work has shown that the photon energy is not very important in the single-shot switching process in MRG [Ref 12], so does the photon energy play an important role here?
1.55 eV (within $\pm$ 0.5 eV) is the photon energy that is typically used in experiments (e.g. [A. V. Kimel et al., Nature 435, 7042 (2005)]). We performed additional simulations for one of the best cases (namely, with $300\,\mathrm{fs}$ width and a large laser intensity of $E_0 = 9.7 \times 10^9\,\mathrm{V\,m^{-1}}$ that is featured in Fig. 4 of the manuscript) consider-

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ing different photon energies as shown in Figure 1. We note that $1.55\,\mathrm{eV}$ is the ideal value that resulted in the largest demagnetization which lead to switching. Increasing or decreasing this value by about $0.5\,\mathrm{eV}$ reduces the demagnetization.

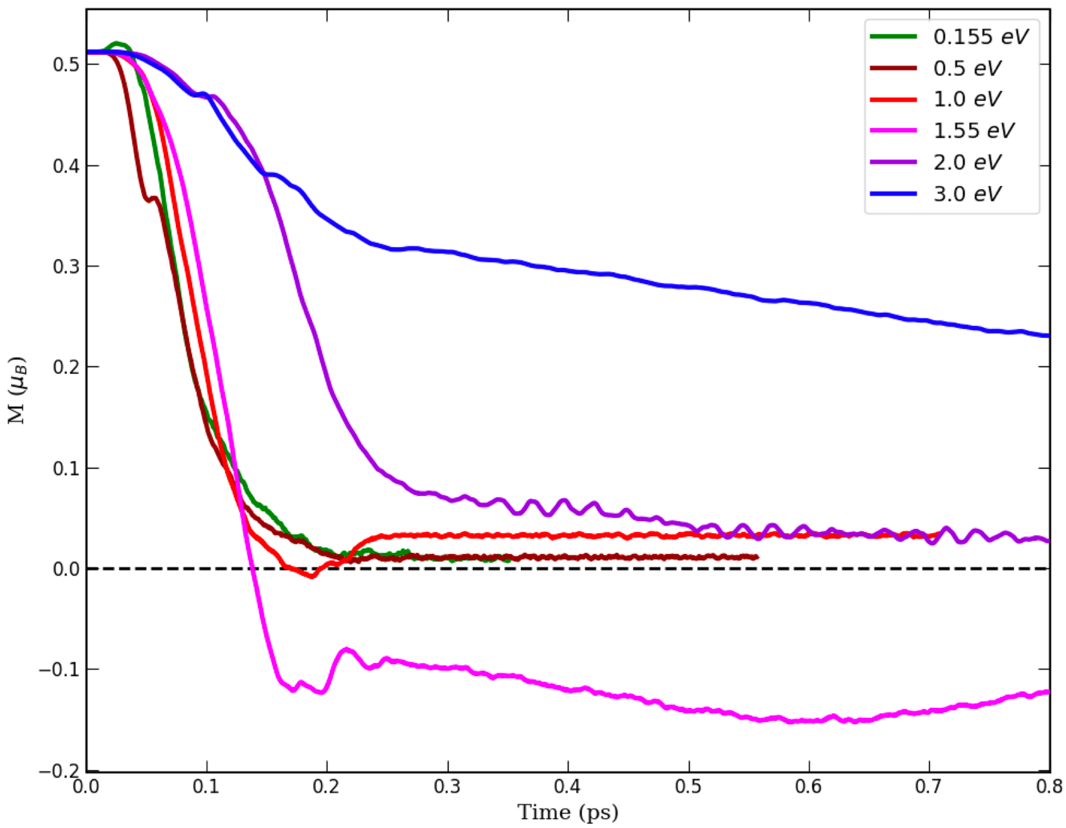

Figure 1: Effect of laser frequency on the magnetization curves in bulk Ni. The pulse used has a width of 300 fs and intensity of $E_0 = 9.7 \times 10^9 Vm^{-1}$.

After addressing all comments and questions of the Reviewer, we hope that she/he is convinced about the importance of having our theoretical manuscript published in Nature Communications Physics.

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Reviewer 3 (Comments to the Author):

The authors of the manuscript "Polarisation-dependent single-pulse ultrafast optical switching of an elementary ferromagnet" use a time-dependent tight-binding implementation to study the real-time dynamics of torque and magnetization in bulk nickel, following laser excitations of different polarization, pulse width, and fluence. From their data they conclude that the magnetization in the material can be switched by a single laser pulse and they report the parameter range for the laser excitation. In addition, by decomposing the results with respect to atom and angular momentum component the authors obtain insight into the underlying mechanism. I find this work interesting and important and I recommend its publication in Communications Physics. Prior to publication, I recommend clarification of the following points:
We thank the reviewer for assessing our manuscript and recommending it for publication.

I highly suggest that that authors comment a bit more on error bars/limitations of their framework. For instance, to what extent does their approach model excitations of the magnetic subsystem? Or are these purely electronic excitations/transitions that are shown and cause the observed behavior? Does this framework allow for fully non-collinear magnetic configurations based on Dirac four vectors, especially during switching?
Our scheme allows for non-collinear magnetic configurations and includes spin-orbit coupling using two-component spinors. As we demonstrate, transient intra-atomic orbital-depenent non-collinear spin-states emerge in the investigated material. So this does not prevent from the possibility of exploring more complex magnetic states. Magnetic excitations are self-consistently described in a time-dependent fashion by inclusion of electron-electron interactions. So far, and as addressed in the manuscript (Discussion part), we neglected dissipation mechanisms induced by the coupling to phonons. Moreover, we did not consider inter-atomic non-collinear magnetic states. Based on the simulations performed by Krieger et al. [J. of chem. theory and computation 11, 4870 (2015)], magnetic non-collinearity within the process of demagnetization from an initial collinear magnetic state is negligible. The work of Chen and Wang [Science Advances 5, eaau8000 (2019)], however, shows that if the initial magnetic state is non-collinear, which could be induced by temperature, the demagnetization magnitude is more important. This aspect is certainly worth investigating in future studies.

In addition, the manuscript, at times, strongly focuses on describing figures, but it is not so clear what physics can be learned or what the underlying takehome message is, e.g. around line 161 on p 11. On line 169, p. 13, the authors mention that the time scale is connected to magnetic anisotropy energy, but do not elaborate
We thank the reviewer for the comment. Since the dynamical simulations that we show are quite rich in information, we decided to point to the important points that we use to draw our conclusions. We also try to add the physical description to the figures themselves (e.g., the vectors and diagrams in Fig. 4 or the second column in Fig. 5). We appreciate that the reviewer carefully read our manuscript and indicated where we missed better explanations. We have improved the take-home message in page 11 to: "The y-component of the torque acts earlier and is larger than the x-component, in line with our interpretation based on the IFE." and in page 13 to: "(...) settled by the non-equilibrium magnetic anisotropy energy. We do not have a precise definition for this quantity, but as the occupations of the different electronic orbitals are strongly modified by the laser we speculate that this leads to a large modification of the effective MAE acting on the magnetic moment.".

Regarding time scales, the comments on lines 203, 212, and 216 can also be made more specific/shown in more detail.
We had the regions marked and named in Fig. 5, but we didn't refer to them in the text. We have now added the references to those regions in the text.

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On line 192, p. 15, the authors discuss that the coupling is "well known", but do not provide a reference.
We have added a reference to the paper [A. Hisazumi, Prog Theor Phys Supp, 101, 11-77 (1990)]; see Table I.

Similarly, wording/clarity can be improved in many places.
With the comments from the reviewers, we hope the new version has improved the overall wording and clarity of the manuscript.

Finally, to strengthen their conclusions, the authors discuss an interesting mechanism around line 199, and it would be very helpful if they could elaborate on its general characteristics and importance. How are the results obtained in this work transferable to other material systems?
The mechanism we describe should be applicable to other materials where the orbitals sp couple antiferromagnetically to the d ones. It should be noted, however, that this is one of several possible mechanisms that may be active in a given material. In layered structures, for example, the spin Hall effect can also generate transverse spin currents which induces a new source of torque in the magnetic layer. For instance, preliminary results on Co/Pt system shows the emergence of dynamical Hall effects and various torques, which we are deeply exploring.

In addition, there are the following minor points:
The authors refer to as "specific algorithm enabling to monitor the non-linear magnetisation dynamics", but seemingly without details?
The real-time propagation is discussed in the Supplementary Note 1 (from Eq. (5) onwards), in particular how we calculate all the time-dependent observables using the embedded-implicit fourth-order Runge-Kutta method. Within this method, both symmetry and energy are preserved, which is necessary for solving Hamiltonian problems. Implicit methods are also more stable compared to explicit ones, especially for stiff problems (i.e., for Differential problems, the ones that have one or more large negative real eigenvalue in their Jacobian [SO. Fatunla. Mathematics of computation 34, no. 150 (1980): 373-390.], there is no exact definition for stiffness though) [J. Chem. Theory Comput. 2018, 14, 6, 3040–3052]. This makes them a good choice for integration over long times, even though being computationally more expensive. Moreover, embedded schemes allow for a low computational cost for step-size control, which becomes useful with weaker perturbations (i.e. the algorithm automatically increases the step size when the error is small). We have added more details about this method on the Supplementary Note 1.

The authors also claim that their approach enables them "to use larger pulse widths". Details are needed. Larger relative to what?
Within our method (see previous point), the time evolution of the wavefunctions is stable up to a few ps, which enables the use of pulses even up to 1 ps and still be able to follow the dynamics for a substantial time interval after the pulse is over. To give an idea to the referee regarding the computational cost of our simulations, a one-picosecond simulation takes one hour on 35 nodes of the JURECA-DC supercomputer in Jülich. We have changed the referred sentence to: "Within our method, the time evolution of the wavefunctions is stable up to a few ps, enabling us to use large pulse widths (up to 1 ps) to investigate the effect of both linearly and circularly polarised pulses over a long time scale (see Methods section for more details). "

On line 123 of page 9, the authors mention a minimum switching threshold that is reasonable and expected,

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but they do not elaborate on why this is expected and what exactly is expected.
The text needed indeed a better explanation. We have changed the text to: The requirement of a minimum intensity threshold for switching is a reasonable and expected condition, since weak laser fields would not excite enough electrons to trigger a strong enough dynamics to induce the switching.

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

The authors have successfully addressed my comments and questions, and revised the manuscript accordingly. I recommend the acceptance of the manuscript.

Reviewer #2 (Remarks to the Author):

The authors have responded somewhat to the referees's concerns, but unfortunately I stand by my original assessment and cannot recommend this paper to be published. The authors appear to have misinterpreted my comments to mean that I (and Reviewer A) support publication in Nature Communications Physics.

My original review actually was intended to suggest that IF the authors can experimentally prove that Nickel's magnetization can be switched, this work should be published in high-impact and prestigious journals. However, extraordinary claims requires extraordinary proof. Without this experimental proof, I can only support publication of this article in e.g. Phys Rev B.

The authors do not provide this proof, which would be rather straightforward to obtain as I explained before, and therefore I am not yet convinced that this work is grounded in reality.

The conflict with experimental results is illustrated, for example, by the authors showing that switching can be obtained using 300fs-long pulses. Experimentally, helicity-dependent all-optical switching in e.g. CoPt is optimized for pulses which are of duration several picoseconds (see e.g. Fig. 1 in Phys. Rev. B 96, 224421 (2017)). Using 250fs-long pulses, the switching cannot be obtained experimentally. It is not clear to me if the mechanism is different or not, and the authors admit that this is not yet understood.

Reviewer #3 (Remarks to the Author):

The authors answered all my questions satisfactorily and I am still quite positive about this manuscript, recommending it for publication.

As a word of caution, I am a bit concerned about using "inverse Faraday effect" as an "umbrella term", because the nomenclature of the different magneto-optical effects and their specific properties/implications/definition never seize to puzzle me. That being said, I think I am fine with it in the present context, due to the preceding explanation of the authors.

Also, while the depth of the insight can always be improved (see response to reviewer #2), I do think that this work provides a crucial, timely, and important result that could encourage future experimental verification/testing and, as such, I view this manuscript as an important contribution.