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Ab Initio Study of Helicity-Dependent Light-Induced Demagnetization: From the Optical Regime to the Extreme Ultraviolet Regime

Philippe Scheid,* Sangeeta Sharma, Gregory Malinowski, Stéphane Mangin, and Sébastien Lebègue

ABSTRACT: We use ab initio real-time time-dependent density functional theory to investigate the effect of optical and extreme ultraviolet (XUV) circularly polarized femtosecond pulses on the magnetization dynamics of ferromagnetic materials. We demonstrate that the light induces a helicity-dependent reduction of the magnitude of the magnetization. In the XUV regime, where the 3p semicore states are involved, a larger helicity dependence persisting even after the passage of light is exhibited. Finally, we were able to separate the part of the helicity-dependent dynamics due to the absorption from the part due to the inverse Faraday effect. Doing so, we show that the former has, overall, a greater impact on the magnetization than the latter, especially after the pulse and in the XUV regime. This work hints at the yet experimentally unexplored territory of the XUV light-induced helicity-dependent dynamics, which, according to our prediction, could magnify the helicity-dependent dynamics already exhibited in the optical regime.

KEYWORDS: ab initio calculations, time-dependent density functional theory, ultrafast phenomena, magnetization, helicity-dependent dynamics

To face the continuing demand for large-density and energy-efficient data storage devices, the possibilities of manipulating the magnetization without relying on the generation of a magnetic field are widely being investigated.1 One of the most promising methods is the use of femtosecond light pulses. Triggered by Beaurepaire et al.,2 this field of study gained a pronounced interest with the discovery of Kimel et al.3 that the light-induced magnetization dynamics of an oxide can be helicity dependent. An even more dramatic phenomenon, named the all-optical helicity-dependent switching (AO-HDS), was evidenced in many different types of magnetic thin films the following years.4–7 AO-HDS allows for the control of the magnetization state by solely relying on circularly left (σ−) or right (σ+) polarized light pulses of a few tens of femtoseconds and, as a consequence, is a prime candidate for the disruption of data storage technologies. While the origin of the AO-HDS is still debated,8,9 it is assumed that the dominating contribution comes from the inverse Faraday effect (IFE), originally predicted by Pitaevskii10 in a non dissipative and nonmagnetic medium. He found that circularly polarized light induces a magnetization proportional to the intensity of the light source at any time, and as a consequence, its contribution to the magnetization dynamics vanishes with the intensity of the light. This effect was recently revisited11,12 and evaluated in different absorptive compounds13 using ab initio means. Another effect, occurring simultaneously to the IFE in dissipative materials, is the magnetization induced during light absorption14 (MILA), whose magnitude is proportional to the fluence of the light. Consequently, and very differently from the IFE, its impact on the magnetization persists even after the presence of the light pulse.

Recently, real-time time-dependent density functional theory (RT-TDDFT) has been successfully applied to describe the linearly polarized laser-induced demagnetization,15 and predicted a new phenomenon occurring during the early dynamics based on optical intersite spin transfer,16–18 which has later been experimentally confirmed.19–21 In the present work, we investigate the dynamics of the magnetization density under the influence of circularly polarized pump pulses using RT-TDDFT. The main aim of the work is to study the dependence of spin dynamics on the helicity of the pump pulse in transition metals (TMs) fcc Ni, fcc Co, and bcc Fe. To this end, we investigate the effect of left and right circularly polarized light of two different kinds, (a) optical pulses and (b) XUV pulses, that resonate with the 3p states of Fe, Co, and Ni. We find that, in both cases, the induced spin dynamics is helicity dependent and that this helicity dependence is larger...
when using a XUV light. The origin of this helicity dependence is closely assessed by examining the spin-resolved time-dependent density of states. Then, we separate the contribution of the IFE and the MILA in the helicity-dependent part of the magnetization dynamics and show that the latter has a greater impact, especially in the XUV regime and when the light pulse vanishes.

The calculations are performed in two steps: first the ground state of the system is calculated using density functional theory.\textsuperscript{13,14} RT-TDDFT is then used to time propagate this ground state under the influence of a pump pulse which is treated as an external vector potential. These calculations are performed with the noncollinear full potential linearized augmented-plane-wave method,\textsuperscript{23} including spin–orbit coupling, as implemented in the ELK code (\texttt{http://elk.sourceforge.net/}). The Brillouin zone is sampled with a 20 × 20 × 20 mesh for each system, and states up to 90 eV above the Fermi energy are included. Within RT-TDDFT,\textsuperscript{28} the dynamics of the Kohn–Sham spinors is evaluated using

\[
\frac{d\psi_{\alpha}(r,t)}{dt} = \left( -\frac{1}{2}\nabla^2 + \frac{i}{\varepsilon}A_{\text{ext}}(t) \cdot \nabla + V_{\text{H}}(r,t) + \frac{1}{2c}\sigma B_{\text{ext}}(r,t) \right)\psi_{\alpha}(r,t) - \frac{1}{4c}\sigma \left(V_{\text{H}}(r,t) \times \nabla V_{\text{H}}(r,t)\right)\psi_{\alpha}(r,t) \tag{1}
\]

In this equation \(\psi_{\alpha}(r,t) = \langle \text{nk}(t) \rangle\) is a two-component spinor valued Bloch orbital indexed by \(k\), a vector of the first Brillouin zone and \(n\) the band index; \(A_{\text{ext}}\) is the vector potential of the light; \(\sigma\) are the Pauli matrices; \(B_{\text{ext}}(r,t)\) is the exchange correlation magnetic field; and \(V_{\text{H}}(r,t)\) is the effective Kohn–Sham potential. The latter is the sum of the external potential, \(V_{\text{ext}}(r,t)\), the Hartree potential, \(V_{\text{H}}(r,t)\), and the exchange correlation potential, \(V_{\text{xc}}(r,t)\). To compute the exchange correlation potential, we use the noncollinear\textsuperscript{27} adiabatic local spin density approximation. The last term of the right-hand side of eq 1 is the spin–orbit coupling. As shown in previous works,\textsuperscript{13,14} its presence is required in order for the magnetization to be affected by the light. While, as discussed by Mondal et al.,\textsuperscript{28,29} it is not the only relativistic contribution appearing up to the second-order expansion of the Dirac Hamiltonian, its contribution to the dynamics is far greater than the others,\textsuperscript{14} which are therefore safely left out here. The magnetization is treated as a fully unconstrained vector field, and the magnetic moment is evaluated using

\[
M(t) = \mu_0 \sum_x \frac{1}{\Omega_bz} \int_{\Omega_bz} \text{d}k |f_{\text{nk}}(t)|^2 \text{d}(\varepsilon_{\text{nk}} - \varepsilon) \tag{2}
\]

where \(f_{\text{nk}}\) is the occupation of the state \(\text{nk}\) defined by the initial state of the Kohn–Sham system; \(\mu_0\) is the Bohr magneton; and \(\Omega_bz\) is the volume of the first Brillouin zone.

The last term in eq 1 is the spin–orbit coupling. Without it, \(\sigma\) would commute with the Hamiltonian, and the magnetization would remain constant during the dynamics, as shown in ref 15. The time propagation is done using the algorithm detailed in ref 17 with a time step of 2.4 fs.

The dynamics of the charge and spin density of bcc Fe, fcc Co, and fcc Ni are evaluated under the action of circularly polarized laser pulses having a Gaussian intensity profile of fwhm = 25 fs and an incident fluence of 13.11 mJ/cm\(^2\). Simulations were performed with an optical pulse of frequency of 1.55 eV and XUV pulses resonating with the lowest-lying 3p states, respectively, located at 55, 60, and 65 eV below the Fermi energy in bcc Fe, fcc Co, and fcc Ni. The pump pulse propagates along the direction of the average magnetic moment, which we define as the \(z\)-axis.

Both Figure 1(a) and (b) show that, as for linearly polarized pump pulses,\textsuperscript{28} circularly polarized pump pulses also lead to spin–orbit coupling mediated\textsuperscript{14,30} demagnetization of TMs. We find that for a fixed incident fluence the demagnetization is more pronounced when using an optical pump pulse, rather than a XUV one. The helicity-dependent magnetization dynamics can also be clearly seen in Figure 1; for an optical pulse, with the exception of Ni, demagnetization induced by \(\sigma^-\) is larger than \(\sigma^+\). This particular behavior of Ni was also noted in ref 14. For XUV pulses, \(\sigma^-\) always leads to a larger demagnetization than its counterpart for all of the studied materials. Also, the difference between the demagnetization from two helicities is much more pronounced for XUV pulses as compared to optical pulses.

These results pose two interesting questions: (i) Why larger demagnetization is produced by optical as compared to XUV pulses? (ii) Why the helicity dependence is more pronounced for XUV pulses than for optical pulses? In order to understand this, we now proceed to the computation of the transient spin-resolved occupied density of states (oDOS) defined as\textsuperscript{18}

\[
oDOS^\sigma(\varepsilon, t) = \frac{1}{\Omega_{bz}} \sum_x \int_{\Omega_{bz}} \text{d}k |f_{\text{nk}}(t)\langle \text{nk}\rangle |^2 \delta(\varepsilon_{\text{nk}} - \varepsilon) \tag{3}
\]

where \(\varepsilon\) is the energy; \(t\) is the time; \(\varepsilon_{\text{nk}}\) is the eigenvalue associated with the Bloch eigenstate \(\text{nk}\); and \(\sigma\) is indexing the spin state, which for a collinear system would be \(\uparrow\) or \(\downarrow\). Finally \(f_{\text{nk}}(t) = \sum_\varepsilon |f_{\text{nk}}(t)|^2\delta(\varepsilon_{\text{nk}} - \varepsilon)\)

Figure 2(a–d) show the changes in oDOS for bcc Fe; i.e., oDOS\(^\sigma(t = 32 \text{ fs}) - oDOS^\sigma(t = 0 \text{ fs})\), for a \(\sigma^+\) and a \(\sigma^-\) polarization of an optical and a XUV pulse. In both cases, this quantity decreases below the ground-state Fermi level (0 eV) and increases above it; i.e., electrons are getting excited and start occupying empty states. Excitations at energies well below 1.55 eV show the presence of nonlinear processes. Comparing Figure 2(a) and (b) with (d) and (e), question (i) is immediately answered: the magnitude of change in the oDOS is almost 25 times larger for both helicities of the optical light, as opposed to the XUV one; i.e., the optical pulse is more absorbed than the XUV pulse. Consequently, and as explained in ref 14, this leads to a larger number of excited electrons.
\[
\zeta(t) = \frac{|M_{\text{HD}}(t)|}{M_{\text{HI}}(0) - M_{\text{HI}}(t)}
\]  

representing the relative magnitude of the HD effect over the HI one (both \(M_{\text{HD}}(t)\) and \(M_{\text{HI}}(t)\) have been convoluted with a Gaussian having a standard deviation of 2 fs). Figure 3(b) and (d) are plots of \(\zeta\) as a function of time for both types of pulses. They evidence that HD effects are more than twice as large in the XUV range than at 1.55 eV. Moreover, while in the optical regime \(\zeta\) tends to be attenuated by the complex dynamics of the interacting electronic density, the light-induced helicity dependence is maintained over a longer course of time in the XUV regime.

In the past, there has been discussion about the factors that lead to HD dynamics. The two main candidates have been identified to be the IFE and the MILA. This past analysis has been done assuming that the magnetic response of the system is dominated by linear order in perturbation. Even though, as mentioned above, our work also includes a nonlinear effect, in order to comment on the relative importance of these contributions, we perform the following analysis: according to Battiato et al.\(^{11,12}\) \(M_{\text{HD}}(t)\) is composed of a transient signal, oscillating parts, the absorptive contribution which leads to MILA\(^{13}\) and the zeroth harmonic which leads to the IFE.\(^{10}\)

To remove the oscillatory and transient contributions which together average to 0, we perform a convolution of \(M_{\text{HD}}\) with a Gaussian having a root-mean-square of 2 fs. This allows us to decompose the HD part of the magnetization dynamics into

\[
M_{\text{HD}}(t) \approx M_{\text{I}}(t) + M_{\text{F}}(t) = \chi I(t) + \gamma_6 \int_0^t dt' I(t')
\]  

where \(I(t)\) is the intensity of the light; and \(\chi\) and \(\gamma_6\) are, respectively, characterizing the magnitude of the HD effect proportional to the intensity, such as the zeroth harmonic generation, and to the fluence, such as the absorption. As presented by Battiato et al.\(^{11}\) and as seen in the results discussed below, the magnitude of both \(\chi\) and \(\gamma_6\) depends on the distribution of frequencies present in the laser pulse. In this framework, the contribution of both effects in \(M_{\text{HD}}\) is uniquely defined by

\begin{figure}
\centering
\begin{subfigure}{0.45\textwidth}
\includegraphics[width=\textwidth]{figure2a.png}
\caption{(a–e) Variation of the spin- and occupation-resolved density of states, \(\sigma\text{DOS}^\sigma(t = 32\, \text{fs}) - \sigma\text{DOS}^\sigma(t = 0\, \text{fs})\), in bcc Fe, induced by a \(\sigma^+\) (a) and (d) and a \(\sigma^-\) (b) and (e) polarization of the light induced after 32 fs using an optical (a and b) and a XUV light (d and e). To improve readability, the \(\downarrow\) curves are multiplied by \(-1\). (c, f) Difference of the total density of states induced by a \(\sigma^+\) and a \(\sigma^-\) polarized light, \(\sigma\text{DOS}_{\text{hel.}} = \sigma\text{DOS}^\sigma_1(t = 32\, \text{fs}) - \sigma\text{DOS}^\sigma_2(t = 32\, \text{fs})\), after 32 fs computed at 1.55 eV (in the XUV range). The right side of (d), (e), and (f) is magnified by a factor of 10.
\end{subfigure}
\begin{subfigure}{0.45\textwidth}
\includegraphics[width=\textwidth]{figure3.png}
\caption{Magnitude of the helicity-dependent dynamics over the helicity-independent one, \(\zeta\), at 1.55 eV (a) and in the XUV range (b).
\end{subfigure}
\end{figure}
The individual contributions of $M_I(t)$ and $M_F(t)$ to the total magnetization density are shown in Figure 4 for bcc Fe, fcc Co, and fcc Ni.

In the case of the optical pump pulse (Figure 4(a)), the magnitudes of both effects (MILA and IFE) are similar during the pulse. The part of the HD dynamics due to the absorption, $M_F$, can be compared to some earlier work\textsuperscript{14} using standard density functional theory, and as seen in the inset of Figure 4(a), we find that at the early stages of the dynamics, where the linear regime is dominant, both the signs and the magnitudes are in agreement.

In the XUV range (Figure 4(b)), the dynamics is dominated by the absorption (MILA), i.e., $M_{HD} \approx M_F$. Consequently, the helicity-dependent magnetization induced by the XUV light will mostly persist even after its intensity has decreased and a large $\zeta$ is obtained at all times, in comparison with the optical excitation (see Figure 3).

In conclusion, using real-time time-dependent density functional theory, we evidenced the presence of a helicity-dependent magnetization dynamics in fcc Ni, fcc Co, and bcc Fe characterized by a difference in the reduction of the magnetic moment carried by the atoms. We found that this helicity dependence is particularly strong when exciting the more localized 3p states. Indeed, by closely examining the time-dependent density of states, we found that the helicity dependence of the dynamics is enhanced for localized states. Then, we disentangled the role of the two main first-order effects which may be held responsible for the helicity dependence, i.e., the inverse Faraday effect and the angular momentum absorption effect. We found that the latter has a far greater impact than the former on the overall dynamics, especially in the XUV range and after the light is gone.

These results also have strong experimental implications and may be responsible for the helicity-dependent demagnetization evidenced by Tsema et al.\textsuperscript{32} and Quessab et al.\textsuperscript{33} Furthermore, the dynamics reported here may also play a role in the AO-HDS\textsuperscript{37} and the helicity-dependent domain wall motion.\textsuperscript{34} Finally, our results are a call for experimental exploration, as they suggest a large enhancement of the efficiency of all of the aforementioned phenomena by going out of the optical range and, in doing so, involving localized core and semicore states.

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Notes
The authors declare no competing financial interest.
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REFERENCES