Coherent optical control of magnons: \textit{ab-initio} predictions

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We demonstrate optical control of magnons using femtosecond laser pulses by performing \textit{ab-initio} real-time TDDFT simulations. We predict that the spin-wave dynamics in Fe\textsubscript{50}Ni\textsubscript{50} can be manipulated by tailoring the applied laser pulse via three distinct mechanisms: (1) element selective destruction of magnon modes depending on the laser intensity, (2) delay dependent freezing of the magnon mode into a transient non-collinear state, and (3) OISTR-driven renormalization of the optical magnon frequency. Harnessing such processes would significantly speed up Magnonic devices.

The next-generation of technology requires non-volatility, faster data processing, decreased power consumption, and increased integration densities. In this regard Spintronics [1–3], an alternative to conventional electronics, is emerging as a powerful candidate. It utilizes the intrinsic quantum-mechanical property of electrons, namely spin, to achieve efficient information encoding. A shining example of this is the Giant Magnetoresistance (GMR) [4, 5] effect which has greatly increased the storage capacity of hard drives. A key component to realizing the full potential of spintronics is Magnonics [6–8] where low-energy (10-1000 meV) spin-wave excitations, in a ferro- or anti-ferro- magnetic materials, are used to carry a pure spin current for long distances (nano-meters). This is much more efficient than regular spin current where energy loss caused by Joule-heating is a limiting factor. While energy efficient, such devices are slow in that the operational times are of the order of a few 100s of picoseconds. In order for faster data processing additional means of spin control need to be developed.

Ultrashort laser technology has emerged as a promising tool for manipulating spins on femtosecond timescales, i.e. several orders of magnitude faster than currently available devices. This is an active and rapidly evolving field of research in which several key processes for ultra-fast spin control have emerged—ultrafast demagnetization [9] whereby loss of spin (or magnetic moment) occurs in less than 100 fs when acted upon by an optical laser pulse, all-optical switching[10][11] in which the spins switch by 180° when excited by the laser, spin transfer from one magnetic sub-lattice to another by charge-excitations induced by the laser, known as Optical Inter-sublattice Spin Transfer (OISTR)[12][13] etc. Harnessing these techniques to control Magnonics would lead to operational times in the femtosecond regime.

In this work we combine Magnonics with one of these ultrafast spin control processes, namely OISTR, to obtain a three fold coherent control of magnons using ultrashort laser pulses— we demonstrate that laser pulses can be tuned to obtain (1) element selective destruction of magnon modes in multi-component magnetic materials, (2) frequency change of selected magnon modes and (3) element selective canting of the magnetic moment i.e. laser induced transient non-collinear state of a ferro-magnet by magnon destruction. OISTTR is a coherent process which is possible in any multi-sub-lattice magnet and has already been demonstrated experimentally for a wide range of materials and geometries, such as bulk Heusler compounds [14], Co/Cu interfaces [15], and Ni/Pt multilayers [16]. As these excitations are directly induced by the pump laser, they take place on the timescale of a few femtoseconds and can be employed as a tool for coherent manipulation of magnons. In order to demonstrate this we use Fe\textsubscript{50}Ni\textsubscript{50} alloy which is a multi-sublattice ferro-magnet and is known[17, 18] to have element specific magnetization dynamics making it a good candidate for the present study.

The usual theoretical approach to study magnons is model based e.g. the Heisenberg model [19] or the Landau-Lifshitz-Gilbert equation [20][22]. These approaches are based on parameterized exchange interactions and assume that the system is in its ground-state and the applied perturbation is very small. In such a sit-
uation the exchange parameters can be extracted from ab-initio DFT calculations. In the present work we plan to study the magnetic excitations in systems which have been knocked out-of-equilibrium by a strong pump-laser pulse, making these approaches inappropriate. Thus we employ a formally exact method for treating the dynamics of magnetization density under external perturbation \[23–26\], namely time-dependent density functional theory (TDDFT) \[27–29\]. TDDFT has previously been used to predict magnon frequencies and lifetimes in the linear-response \[30–32\] regime, however in the present work we extend it to probe, in real-time, the magnons and the effect of non-linear perturbation on magnon modes. The fundamental quantities in TDDFT are the density and the magnetization density which are obtained by Fourier transforming the transverse moments. TDDFT is formally exact state-of-the-art method for treating Magnonics in out-of-equilibrium systems, but the price to pay for such a treatment is that it is highly computationally demanding. This restricts the size of supercell that is practical and hence limits our study to high energy/high \(q\) modes. However, the physics of the problem remains valid even at lower \(q\) values.

Since one of the main aims of this work is to be able to element selectively manipulate the magnons, we begin by addressing the question are there any decoupled modes in the system, i.e. modes where the two magnetic sub-lattices oscillate independently? We study magnons in a 4 atom super-cell which is formed by extending the L10 primitive cell along the c-axis with lattice parameters \(a = 3.85\AA\) and \(c = 7.71\AA\). The Brillouin zone was sampled on a \(k\)-grid of \(8 \times 8 \times 8\) and a time step of 1.209 attoseconds was used for time-propagating the orbitals using the algorithm presented in Ref. \[35\]. The adiabatic local spin density approximation to the XC functional was used. All simulations were done using the all-electron ELK electronic structure code \[36\].

![FIG. 2: Oscillation of the transverse (x,y) magnetic moments of the individual nickel and iron atoms in a 4-atom supercell of Fe50Ni50 for different initial states. These magnons correspond to momenta \(q = \Gamma, \pm 1/2 \Gamma X\), and X. Decoupled, element specific magnon modes can be seen for (a) nickel and (b) iron. Coupled Goldstone and optical modes can be seen in (c) and all 4 modes are excited in (d).](image)

This 4 atom supercell of Fe50Ni50 allows four magnon modes with wave-vectors \(\Gamma, \pm 1/2 \Gamma X\), and \(X\) where \(X = (0, 0, 2\pi/a)\). TDDFT calculations reproduce these modes and they are shown in Fig. 1. Two modes out of these four are decoupled in that only one of the magnetic sub-lattices oscillates: 1) high energy pure Ni mode with \(\omega = 710\) meV (Fig. 1(a)). The energy of this mode is higher than the corresponding mode in bulk Ni (390 meV). 2) A low energy pure Fe mode with \(\omega = 90\) meV (see Fig. 1(b)), the frequency of this mode is also higher than the corresponding mode in bulk Fe (65 meV). The reason for existence of these decoupled modes is the fact that at
wave-vector \( q = \pm 1/2 \Gamma X \), the effective exchange fields acting on an atom, from nearest-neighbor atoms of the other species, cancel leading to only one of the magnetic sub-lattices to oscillate. The other two modes, out of the four allowed modes, are the coupled Fe and Ni modes: 3) the Goldstone mode \( (\omega = 0) \) whereby all spins tilt together, as seen in Fig. 3 (c) dotted lines and 4) the optical mode, where the Fe and Ni oscillate 180° out-of-phase with each other, as can also be seen in Fig. 3 (c) full lines. The frequency of this mode is 760 meV, much higher than the \( q = X \) mode in either Fe or Ni. All these modes can also be excited at the same time, as in Fig. 2 (d).

Element selective optical destruction of magnons: In order to be able to manipulate magnons at ultra-fast time scales we now investigate the behavior of magnon modes under short laser pulses. One of the fastest possible spin response to lasers is via OISTR, primarily driven by minority spin electrons optically excited from one magnetic sub-lattice to another, causing an increase in the moment on the first sub-lattice. In the present work the materials, as well as the laser pulses, are chosen to maximize OISTR: in the Fe\(_{50}\)Ni\(_{50}\) alloy the magnetic moment on the Fe sub-lattice (2.88 \( \mu_B \)) is much higher than on the Ni sub-lattice (0.64 \( \mu_B \)). This causes laser induced optical excitations to transfer minority spin electrons from Ni to Fe, which in turn leads to an increase in the moment on the Ni site, while a corresponding decrease on the Fe site (see Fig. 3 (b)). The frequency of the laser pulse (2.19 eV) is tuned to optimize this charge transfer. The effect of OISTR on magnon modes can be seen in Fig. 3– strong laser pulse (incident fluences of 9.6807 mJ/cm\(^2\) and FWHM of 2.41 fs) effectively destroy both decoupled modes (see Fig. 3 (d)); the amplitude of the pure Fe magnon mode collapses with only small oscillations remaining which are also quickly damped. Looking at the Ni moments, which initially are a superposition of the pure Ni mode and the optical mode, we see that now only the optical mode exists as the two Ni atoms behave identically (recall that in the pure Ni mode, the two are 180° out-of-phase).

These magnon modes show a different dynamics when subjected to a weaker laser pulse of incident fluence 0.9537 mJ/cm\(^2\); the pure nickel mode now survives while the Fe mode is still destroyed (see in Fig. 3 (e)). In this case the Fe atoms cant with respect to each other with a new, but much reduced, pure Fe mode oscillating about this new configuration. By examining the amount of spin up/down electrons excited on each atom, we find that the Fe atoms have significantly more local optical excitations than Ni. This causes the Fe-Fe exchange coupling to be modified more strongly than the Ni-Ni coupling, explaining the difference in behavior between the two modes. Thus we have found a method by which we can selectively destroy either both Fe and Ni modes or just the Fe mode, on a femtosecond timescale by tuning the fluence of the laser pulse. This is an important finding as it not only offers a mechanism of control over magnons but also highlights the fact that the dynamics of element specific magnetization in alloys can greatly differ due to choice of pump pulse [11, 17].

Control of canting vector: Change in relative directions of the moments (i.e. angle between the inter-site spins...
FIG. 4: The canting vector is dependent on the delay time of the laser and is shown relative to the pure Fe mode oscillations. A laser of fluence 0.9537 mJ/cm$^2$ is applied at (a) 16.8 fs and (b) 8.4 fs on the pure iron mode.

in a multi-sub-lattice system) in a material can be obtained by rapid destruction of selected magnon modes. We demonstrate, in Fig. 4, that the canting vector, $\mathbf{m}^{Fe_1}(t) - \mathbf{m}^{Fe_2}(t)$, can be controlled using the time delay of the laser pulse. The effect of the laser pulse is to destroy the magnon mode, but the phase of the magnon mode at the point when the laser is applied determines in which direction the Fe moments eventually point, and thus determines the direction of the canting vector. In the first scenario the center of the pulse is chosen to be located at 16.8 fs when the Fe$_1y$ and Fe$_2y$ are at their maximum amplitude (see Fig. 4 (a)) and in the second case the center of the pulse is chosen to be at 8.4 fs which corresponds to the point in time when the Fe$_1x$ and Fe$_2x$ moments are at their maximum amplitude (see Fig. 4 (b)). These two delays in the laser pulses result in different directions of the canting vector. The laser pulse used to obtain this canting is very weak with fluence=0.9537 mJ/cm$^2$. Unlike in the strong laser case of Fig. 3 (d), where the amplitude of the Fe mode is reduced to zero, in Fig. 4 the Fe moments remain finite but cease precessing, resulting in a final canted transient state. The main reason behind this is that the laser excitation disrupts the exchange coupling between the nearest Fe atoms, causing the magnon mode to freeze into a spin spiral configuration. Extending the delay by half a period of the magnon oscillation will result in a canting vector pointing in opposite direction. This indicates that with a careful choice of laser pulse a ferromagnetic metal can be made to be transiently non-collinear with a certain degree of control over the angle between inter-site spins.

**Ultrafast change in magnon frequency:** The frequency of the magnon modes can also be manipulated by pump-laser pulse. To demonstrate this we excite the optical mode and then look at its dynamics under the influence of pump pulses of differing fluences. The results, for two different laser intensities (0.9537 and 9.68 mJ/cm$^2$) are shown in Figs. 5 (a) and (b) where it is clear that the oscillations are strongly influenced by the laser. Fourier transform of the transverse moments during these oscillations gives the frequency of the magnon mode and this is plotted, as function of laser intensity, in Fig. 5(c). The main reason behind this change in frequency is the weakened exchange field between the magnetic sub-lattices due to two processes, both of which lead to increased screening – (a) excitation of electrons to excited delocalized states and (b) transfer of localized charge from one atom to the other. This implies that the stronger this charge transfer is, the greater the change in the magnon frequency, a fact that is reflected in the linear dependence of the magnon frequency on the pump-pulse fluence (Fig. 5 (c)). At some higher intensity where the charge excitation process saturates, so would the change in the magnon frequency. Thus optical excitations offer a direct control of frequency of a coupled magnon mode of two sub-lattices via tuning of the fluence of the laser pulse. Since OISTR effects are very strong on AFM coupled systems, we expect very large changes in magnon modes when pumped with lasers.

In conclusion, we have extended the domain of TDDFT simulations to include magnon dynamics in real-time. This opens the field of laser-coupled Magnonics to ab-initio theory. We first showed the prediction by TDDFT of element specific magnon modes with vastly different energies in Fe$_{50}$Ni$_{50}$ alloy. We then demonstrated three ways in which ultrafast laser pulses can control magnon dynamics: (1) selective destruction of particular magnon modes where the Ni or Fe modes could be selectively destroyed depending on the laser intensity, (2) laser driven destruction of magnon mode leading to transient non-collinear state of a ferro-magnet, and (3) OISTR-driven renormalization of the optical magnon frequency, where we found a linear dependence between the laser intensity (or moment transferred) and the decrease of the magnon frequency. In all cases the outcomes were achieved on ultrafast
timescales thus demonstrating the potential of laser control of Magnonics for future technology. In future work, we plan to study the more exotic magnons in AFM systems and magnetic insulators (which are more long-lived due to the lack of Landau damping).

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[1] S. Bader and S. Parkin, Annual Review of Condensed Matter Physics 1, 71 (2010).
[2] I. Zutić, J. Fabian, and S. Das Sarma, Rev. Mod. Phys. 76, 323 (2004).
[3] V. Baltz, A. Manchon, M. Tsoi, T. Moriyama, T. Ono, and Y. Tserkovnyak, Rev. Mod. Phys. 90, 015005 (2018).
[4] G. Binasch, P. Grünberg, F. Saurenbach, and W. Zinn, Phys. Rev. B 39, 4828 (1989).
[5] M. N. Baibich, J. M. Broto, A. Fert, F. N. Van Dau, P. Petroff, E. P. Etienne, G. Creuzet, A. Friederich, and J. Chazelas, Phys. Rev. Lett. 61, 2472 (1988).
[6] D. Steil et al.
[7] G. Gubbio, T. Siachi, M. Madami, G. Carlotti, A. O. Adeyeye, and M. Kostylev, Journal of Physics D: Applied Physics 43, 264003 (2010).
[8] V. V. Kruglyak, S. O. Demokritov, and D. Grundler, Journal of Physics D: Applied Physics 43, 260301 (2010).
[9] E. Beaurepaire, J.-C. Merle, A. Daunois, and J.-Y. Bigot, Phys. Rev. Lett. 76, 4250 (1996).
[10] C. D. Stanciu, F. Hansteen, A. V. Kimel, A. Kirilyuk, A. Tsukamoto, A. Itoh, and T. Rasing, Phys. Rev. Lett. 99, 047601 (2007).
[11] I. Radu, K. Vahaplar, C. Stamm, T. Kachel, N. Pontius, H. A. Dürr, T. A. Ostler, R. W. Chantrell, A. Tsukamoto, A. Itoh, A. Kirilyuk, T. Rasing, and A. V. Kimel, Nature, 205 (2011).
[12] P. Elliott, T. Müller, J. K. Dewhurst, S. Sharma, and E. K. U. Gross, Scientific Reports 6 (2016).
[13] J. K. Dewhurst, P. Elliott, S. Shallcross, E. K. U. Gross, and S. Sharma, Nano Letters 18, 1842 (2018).
[14] D. Steil et al., in prep. (2019).
[15] J. Chen, U. Bovensiepen, A. Eschenlohr, T. Müller, P. Elliott, E. K. U. Gross, J. K. Dewhurst, and S. Sharma, Phys. Rev. Lett. 122, 067202 (2019).
[16] P. Siegrist, J. A. Gessner, M. Ossiander, C. Denker, Y. Chang, A. Schroder, M. C. Guggenmos, Y. Cui, U. Walowski, J. A. Clark, and Martin, J. K. Dewhurst, U. Kleineberg, M. Muenzenberg, S. Sharma, and M. Schultz, arXiv:1812.07420 (2019).
[17] S. Mathias, C. La-O-Vorakiat, P. Grychtol, P. Granitzka, E. Turgut, J. M. Shaw, R. Adam, H. T. Nembach, M. E. Siemens, S. Eich, C. M. Schneider, T. J. Silva, M. Aeschlimann, M. M. Murnane, and H. C. Kapteyn, Proceedings of the National Academy of Sciences 109, 4792 (2012).
[18] I. Radu, C. Stamm, A. Eschenlohr, F. Radu, R. Abrudan, K. Vahaplar, T. Kachel, N. Pontius, R. Mitzner, K. Hollbach, A. Föhlsch, T. A. Ostler, J. H. Mintink, R. F. L. Evans, R. W. Chantrell, A. Tsukamoto, A. Itoh, A. Kirilyuk, A. V. Kimel, and T. Rasing, SPIN 05, 1550004 (2015) https://doi.org/10.1142/S2010324715500046.
[19] W. Heisenberg, Zeitschrift für Physik 49, 619 (1928).
[20] R. Mondal, M. Berthiau, and P. M. Oppeneer, Phys. Rev. B 94, 144419 (2016).
[21] D. Hinzke, U. Axtitia, K. Carva, P. Nieves, O. Chubykalo-Fesenko, P. M. Oppeneer, and U. Nowak, Phys. Rev. B 92, 054412 (2015).
[22] R. F. L. Evans, W. J. Fan, P. Chureemart, T. A. Ostler, M. O. A. Ellis, and R. W. Chantrell, Journal of Physics: Condensed Matter 26, 103202 (2014).
[23] J. K. Dewhurst, S. Shallcross, E. K. U. Gross, and S. Sharma, Phys. Rev. Applied 10, 044065 (2018).
[24] K. Krieger, P. Elliott, T. Müller, N. Singh, J. K. Dewhurst, E. K. U. Gross, and S. Sharma, Journal of Physics: Condensed Matter 29, 224001 (2017).
[25] P. Elliott, K. Krieger, J. K. Dewhurst, S. Sharma, and E. K. U. Gross, New Journal of Physics 18, 013014 (2016).
[26] V. Shoeken, M. Sanchez Piaia, J.-Y. Bigot, T. Müller, P. Elliott, J. K. Dewhurst, S. Sharma, and E. K. U. Gross, Phys. Rev. Lett. 119, 107203 (2017).
[27] E. Runge and E. K. U. Gross, Phys. Rev. Lett. 52, 997 (1984).
[28] P. Elliott, F. Furche, and K. Burke, Excited States from Time-Dependent Density Functional Theory, Vol. 26 (Wiley, 2009).
[29] S. Sharma, J. K. Dewhurst, and E. K. U. Gross, "First Principles Approaches to Spectroscopic Properties of Complex Materials", Eds. C. Di Valentin, S. Botti and M. Coccioni, Topics in Current Chemistry 347, 235 (2014).
[30] N. Singh, P. Elliott, T. Nautiyal, J. K. Dewhurst, and S. Sharma, Phys. Rev. B 99, 035151 (2019).
[31] P. Buczek, A. Ernst, and L. M. Sandratskii, Phys. Rev. B 84, 174418 (2011).
[32] S. Y. Savrasov, Phys. Rev. Lett. 81, 174425 (1998).
[33] T. Kampfrath, A. Sell, G. Klatt, A. Pashkin, S. Mährlein, J. Dekorsy, M. Wolf, M. Fiebig, A. Leitenstorfer, and R. Huber, Nature Photonics 5 (2010), 10.1038/nphoton.2010.259.
[34] J. Dewhurst, K. Krieger, S. Sharma, and E. K. U. Gross, Proceedings of the National Academy of Sciences 109, 4792 (2012).
[35] https://www.pnas.org/content/109/13/4792.full.pdf.
[36] P. Elliott, F. Furche, and K. Burke, Excited States from Time-Dependent Density Functional Theory, Vol. 26 (Wiley, 2009).
[37] S. Sharma, J. K. Dewhurst, and E. K. U. Gross, "First Principles Approaches to Spectroscopic Properties of Complex Materials", Eds. C. Di Valentin, S. Botti and M. Coccioni, Topics in Current Chemistry 347, 235 (2014).
[38] P. Elliott, F. Furche, and K. Burke, Excited States from Time-Dependent Density Functional Theory, Vol. 26 (Wiley, 2009).
[39] S. Sharma, J. K. Dewhurst, and E. K. U. Gross, "First Principles Approaches to Spectroscopic Properties of Complex Materials", Eds. C. Di Valentin, S. Botti and M. Coccioni, Topics in Current Chemistry 347, 235 (2014).
[40] N. Singh, P. Elliott, T. Nautiyal, J. K. Dewhurst, and S. Sharma, Phys. Rev. B 99, 035151 (2019).
[41] P. Buczek, A. Ernst, and L. M. Sandratskii, Phys. Rev. B 84, 174418 (2011).
[42] S. Y. Savrasov, Phys. Rev. Lett. 81, 174425 (1998).
[43] T. Kampfrath, A. Sell, G. Klatt, A. Pashkin, S. Mährlein, J. Dekorsy, M. Wolf, M. Fiebig, A. Leitenstorfer, and R. Huber, Nature Photonics 5 (2010), 10.1038/nphoton.2010.259.