Ultrafast control of magnetization by femtosecond laser pulses

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Abstract

I will present the theoretical background of interactions between light and magnetization. I will then describe two experiments where changes of magnetization induced by laser pulses were observed. At the end, I will discuss possibilities of using the phenomenon in practical applications.
1 Introduction

Magneto-optics is a branch of physics that studies various effects arising from the interaction of light with magnetized media. Initially linearly polarized light, after interaction with such materials, can exhibit both ellipticity and a rotation of the polarization state. These effects are generally categorized into two phenomena - the Faraday effect which occurs when electro-magnetic radiation is transmitted through a magnetized media, and the Kerr effect which addresses the reflections from magnetized media.

These phenomena are widely applied in magnetic research today since they are potentially useful as a means of storing informations in increasingly miniaturized magnetic particles. Recently, a new series of experiments has begun investigating the possibilities of spin manipulation with photons.

In the seminar I will address the question of manipulation of spins in a magnetic material with ultrafast laser pulses. I will present the theoretical background of the phenomenon, two ways of manipulating the magnetization (thermal and non-thermal), and the experimental technique used to observe these effects. I will present the results of the first experiment, where changes of magnetization on the sub-picosecond scale [1] were observed, and then describe the results of an experiment that was made to demonstrate that the non-thermal manipulation of the magnetization is also possible [2].

2 Theoretical background

2.1 Faraday effect

The Faraday effect was discovered in 1845 when Michael Faraday noticed a change in the polarization of the light, passing through glass in the magnetic field.

The Faraday effect is a result of breaking of the time-reversal symmetry in the magnetic field. Because of that, light with different circular polarizations propagates with different speeds, and therefore gains different phase shifts. When both beams recombine, this results in the rotation of the polarization plane.

The dielectric tensor can generally be represented as [3]:

$$\epsilon = \begin{pmatrix} \epsilon_{xx} & \epsilon_{xy} & \epsilon_{xz} \\ \epsilon_{yx} & \epsilon_{yy} & \epsilon_{yz} \\ \epsilon_{zx} & \epsilon_{zy} & \epsilon_{zz} \end{pmatrix}$$

(1)

We can further decompose this tensor to a symmetric part (that can be diagonalized by an appropriate rotation and does not contribute to the Faraday effect) and an antisymmetric part. From now on I will assume that
the symmetric part is isotropic with the dielectric constant $\epsilon$. To see the effect of the antisymmetric part, I will consider the following tensor:

$$
e' = \epsilon \begin{pmatrix} 1 & iQ_z & -iQ_y \\
-iQ_z & 1 & iQ_x \\
iQ_y & -iQ_x & 1 \end{pmatrix} \tag{2}$$

The two normal modes are left and right-circularly polarized beams with refraction index $n_L = n(1 - \frac{1}{2} \vec{Q} \cdot \hat{k})$ and $n_R = n(1 + \frac{1}{2} \vec{Q} \cdot \hat{k})$ where $n = \sqrt{\epsilon}$ is the average refraction index and $\vec{Q} = (Q_x, Q_y, Q_z)$ is called the Voigt vector. $\hat{k}$ is the unit vector along the direction of the light propagation. In general, $Q_i$ are wavelength-dependent, but over a narrow range of wavelengths they might be treated as constants. In the most general case of an absorbing MO material $Q_i$ may be complex numbers. The rotation of the polarization plane (\(\theta\)) after the light has traveled a distance $L$ through the medium is then given as:

$$\theta = \frac{\pi L}{\lambda} (n_L - n_R) = -\frac{\pi Ln}{\lambda} \vec{Q} \cdot \hat{k} \tag{3}$$

This can also be represented as

$$\theta = \frac{\chi L}{n} \vec{M} \cdot \hat{k}, \tag{4}$$

where $\chi$ is magneto-optic susceptibility that is a scalar value in isotropic media and $\vec{M}$ is magnetization \[2\].

The other commonly used notation, where the field is oriented along the $z$-axis, is \[4\]:

$$\theta = VB L, \tag{5}$$

where $V$ is the Verdet constant, given as

$$V = \frac{-4\pi^2 \chi}{n_0 \lambda_0}. \tag{6}$$

Because the samples studied are usually thin and the applied magnetic field is around 1 T, the changes of polarization are in order of magnitude of some tenths of degrees. This heavily depends on the material.

### 2.2 Inverse Faraday effect

The Faraday effect can also work in the opposite direction. In that case, a static magnetization $\vec{M}(0)$ is induced by external oscillating electrical field
with the frequency $\omega$, in our case this is achieved with a high intensity laser pulse. Its induced magnetization is proportional to the vector product of $\vec{E}$ and $\vec{E}^*$, given in [2] as:

$$\vec{M}(0) \propto [\vec{E}(\omega) \times \vec{E}^*(\omega)]$$  \hspace{1cm} (7)

From this equation we see that the circularly polarized light with the frequency $\omega$ should induce a magnetization along the wave vector $\vec{k}$. Because $\vec{E}$ is in the vector product, we see that left- and right-handed polarization waves should induce magnetization of opposite signs. This can be explained phenomenologically, we take the lowest order element of the Taylor series that gives us real value (this is why we use $\vec{E}^*$) and is odd according to the direction (this is why we use the vector product). One more reason for the vector product is that the result has to be a vector itself, therefore a scalar product does not come into account.

According to [2], the magnetization induced can be comparable to the saturated magnetization.

### 2.3 Magneto-optic Kerr effect

The magneto-optic Kerr Effect (MOKE) describes changes of the reflections from the magnetized media. Basically, it is similar to the Faraday effect that describes the light passing through the media. MOKE can be further categorized by the direction of the magnetization vector with respect to the reflection surface and the plane of incidence. When the magnetization vector is perpendicular to the reflection surface and parallel to the plane of incidence, the effect is called the polar Kerr effect. To simplify the analysis, near normal incidence is usually employed when doing experiments in the polar geometry. In the longitudinal effect the magnetization vector is parallel to both the reflection surface and the plane of incidence. When the magnetization is perpendicular to the plane of incidence and parallel to the surface it is said to be in the transverse configuration.

Again, the change of polarization is in order of some degrees, depending on the material.

MOKE, Faraday effect and inverse Faraday effect are the most common techniques employed to observe changes in magnetization of a magnetic material on short time scales.

### 3 Pump-probe technique

The technique used in the experiment is called femtosecond spectroscopy. A mode locked pulse laser (usually Ti:Sapphire or Nd:YAC) is generating short laser pulses that are split by a beam-splitter. One of the beams (probe) is led through a delay line and then both beams are focused on a sample. The
powerful laser beam (pump) excites the sample, that is then probed with the weak pulse (probe). The delay line consist of a cube corner reflector mounted on a translator. The length of the delay line can be changed on micrometer scale so the delay between both pulses can be controlled in order of femtoseconds. This technique is usually used to observe changes in reflectivity, absorption, or in the following cases, rotation of polarization. Using it, we can obtain a time-resolved picture of the changes.

4 Thermal demagnetization

The magnetization of a magnetic material usually decreases upon heating. Above the Curie temperature the long range magnetic order is lost and no net magnetization is left. Heating can be performed using intense laser pulses. Using an optical pump-probe technique, this phenomenon was first observed on sub-picosecond scale by Beaupaire et al. [1] in 1995. Figure 3 shows the remanent magnetization of a nickel film, i.e. its spontaneous magnetization in absence of an external field, as a function of the pump-probe delay $\delta t$. At $\delta t = 0$ the magnetization drops sharply, reaching its minimum value already 2 ps after excitation.
When photons are absorbed by the metal, their energy is transferred to the electron system, creating highly energetic electrons (>1 eV). The electrons are excited from occupied states below the Fermi level to empty states above. This creates a non-equilibrium distribution of highly energetic electrons. The relaxation process that follows consists of three different regimes. The events occur on a picosecond scale.

First, the excited electrons thermalize to a Fermi-Dirac distribution because of e-e scattering. Because of this process, the number of excited electrons increases greatly but their average energy is reduced. After the thermalization, the electrons can be described as in equilibrium with the temperature $T_e$. This process is fast and happens on a timescale of 100 fs.

The second regime is the interaction with the lattice system that is initially unaffected by the optical excitation. However, interactions between electrons and phonons (e-p) will equilibrate the lattice temperature ($T_l$) and thus increasing the lattice temperature. In equilibrium, $T_e = T_l$. This process is slower than the electron-electron scattering. Relaxation time is on the order of picoseconds.

The third regime includes absorption of angular momentum by the lattice and is not fully understood yet.

Magnetization can be reduced by two means. First, the net spin moment can be reduced by interactions with the electron system – by spin flip scattering via electron-electron interactions. Microscopically, the interactions are
caused by the spin-orbit coupling:

$$H_{SO} = \lambda \vec{L} \cdot \vec{S} = \lambda [L_z \cdot S_z + \frac{1}{2}(L^+ \cdot S^- + L^- \cdot S^+)], \quad (8)$$

where $\vec{L}$ and $\vec{S}$ are orbital and spin momentum operators and $\lambda$ is a constant describing the strength of interaction. Due to the spin-orbit interaction, the spin-up and spin-down states are mixed. Because of this interaction the spin expectation value can be raised ($S^+$) if simultaneously the orbital momentum is lowered ($L^-$) and vice versa. Here, we neglect the coupling to the lattice and assume that the contribution of the photons to the angular momentum is small. Therefore, the angular momentum $\vec{J} = \vec{L} + \vec{S}$ is conserved. The local magnetic moment of a material is given by:

$$\vec{\mu} = \mu_B (\vec{L} + g \vec{S}), \quad (9)$$

where an electron $g$ factor is approximately 2. Transfer of the momentum from $\vec{S}$ to $\vec{L}$ can then reduce magnetization only for around 50%.

Another possibility would be the electron-magnon scattering where the spin of an electron is changed by creation or absorption of a collective spin excitation, a magnon. However, this excitation carries the change the electron’s spin moment, so the total spin moment is preserved. To further reduce the magnetization, interactions with the lattice are required in order to absorb the angular momentum.

### 4.1 Three temperature model

In [1], Beaurepaire *et al.* used the three temperature model (3TM) to describe the dynamics after thermalization. The model separates three systems (three coupled heat reservoirs) - the electrons, the lattice, and the spins. Each of them has its temperature, $T_e$, $T_l$, and $T_s$ accordingly. The laser pulse interacts only with the electrons and not with the spins or the lattice. Because the three temperatures are not the same since the electrons were put into the excited state, the temperature differences between the three systems act as the driving force for the energy transfer until the system reaches the new equilibrium where all three temperatures are equal. The process can be described with three coupled differential equations:

$$c_e(T_e) \frac{\partial T_e}{\partial t} = -g_{el}(T_e - T_l) - g_{es}(T_e - T_s) + P(t) \quad (10)$$

$$c_l(T_l) \frac{\partial T_l}{\partial t} = -g_{el}(T_e - T_l) - g_{sl}(T_l - T_s) \quad (11)$$

$$c_s(T_s) \frac{\partial T_s}{\partial t} = -g_{es}(T_s - T_e) - g_{sl}(T_s - T_l) \quad (12)$$

where $T_i$ are the temperatures, $c_i$ are the temperature dependent heat capacities, $g_{ij}$ are the coupling constants between the subsystems and $P(t)$ is the laser power.
Beaurepaire et al. calculated all three heat capacities using this model in combination with experimental results. Figure 5 represents the experimental spin ($T_s$) and electron ($T_e$) temperatures on a thin Nickel film (a) and calculated temperatures that follow from equations 10-12.

![Figure 4: Three temperature model scheme [5]](image4)

5 Non-thermal demagnetization

The experiment carried out by A.V. Kimel et al. in 2004 [2] demonstrated that it is also possible to manipulate the magnetization non-thermally. A sample of DyFeO$_3$ was investigated using femtosecond laser pulses in the
pump-probe configuration to get the time-resolved picture of the magnetization changes. It was shown that a circularly polarized laser pulse indeed rapidly induces a change of magnetization in the sample, besides, left and right-handed polarizations exhibit different changes. This behaviour appears due to the inverse Faraday effect.

5.1 DyFeO₃

The studied material was dysprosium orthoferrite DyFeO₃ which belongs to the group of rare-earth orthoferrites. The material crystallizes in an orthorhombic perovskite-type structure with four molecular units per unit cell. The space-group symmetry is $D_{2h}^{20}$ ($Pbnm$). The spins of Dysprosium ions are not ordered above 4 K, being in a paramagnetic state. The spins of Fe$^{3+}$ ions ($3d^5$) are coupled antiferromagnetically by isotropic exchange.

DyFeO₃ was chosen because it exhibits a giant Faraday rotation of about 3,000° cm$^{-1}$ owing to its strong spin-orbit interaction. Thus non-thermal effects of light on the spontaneous magnetization are expected to be significant in this material.

![Figure 6: Structure of DyFeO₃. The unit cell is orthorhombic perovskite-type with four molecular units per unit cell [6]](image)

5.2 Results and interpretation

The research team used amplified 200 fs pulses from a Ti:sapphire laser at a repetition rate of 1 kHz. The pump beam was circularly and the probe beam linearly polarized. The intensity ratio between both pulses was about 100. Both beams were focused on the sample to a spot diameter of about 200 µm for the pump and a bit smaller for the probe. The intensity of the pump beam was around 30 mJcm$^{-2}$. The experiment was carried out in the temperature range 15-300 K.
For the detection of the optically induced magnetization, the direct magneto-optical Faraday effect was used. Figure 7 shows the temporal evolution of the Faraday rotation in the sample for two circularly polarized pump pulses of opposite helicities. On the scale of 60 ps one can clearly distinguish two different processes that start after excitation with a pump pulse. At a zero time delay, instantaneous changes of the Faraday rotation are observed. This can be attributed to the magneto-optic susceptibility change because of the electron excitation. In a few picoseconds, susceptibility stabilizes because the electron relaxation is fast.

![Figure 7: Faraday rotation in DyFeO$_3$ as a function of the delay time. Two lines represent excitations induced by the pulses of different circular polarizations [2].](image)

The graph at the longer delays can be interpreted as a thermal, coupled with a non-thermal response. The thermal response, as discussed in section 4, is coupled with oscillations with a frequency of about 200 GHz. The oscillations of the magnetization are not thermally induced. It is seen from Figure 7 that the helicity of the pump beam controls the sign of the photo-induced magnetization. This observation unambiguously indicates that the coupling between spins and photons in DyFeO$_3$ is direct, because the phase of the spin oscillations is given by the sign of the angular momentum of the exciting photon. Without the thermal effect, both lines would be symmetrical.

The research team further studied temperature dependence of the process. Figure 8 shows the difference between the Faraday rotations induced by right- and left-handed circularly polarized pump light in the sample for
the temperature range between 20 K and 175 K.

Figure 8: The difference between the signals for right- and left-handed circularly polarized pump pulses in DyFeO3 measured at different temperatures in the range between 20K and 170 K. The difference is plotted rather than the signal to exclude the effects that are not relevant to magnetic excitations. Every new curve is shifted from the previous one along the vertical axis over 0.068 for better view. The box on the top shows the amplitude of the spin oscillations as a function of pump fluence. [2]

At lower temperatures, thermal contribution is smaller because there is less absorption. An increase of the temperature also results in an increase of the frequency of the oscillations up to 450 GHz at 175 K, while the amplitude of the oscillation decreases. The damping of the oscillations in the range of 200 ps is due to the magnon scattering on phonons and spins of dysprosium ions. The highest value of the amplitude of the photo-induced oscillations is observed between 20 K and 50 K.

The effect of such a 200 fs laser pulse on the magnetic system is equivalent to the application of a magnetic field pulse of about 5 T. According to the measurements, the absorption in DyFeO3 is low, so it would also be possible to use more powerful pulses without damaging the sample.
6 Applications

The studied phenomenon seems promising in connection with precise methods for storing informations in increasingly miniaturized magnetic particles. It is still a question how to obtain a permanent change of magnetization since the change in the experiment was only in matter of picoseconds.

Traditional magnetic storage media records data bits as a series of small magnetic domains with reversed magnetizations in the plane of the media. Hard disks that are commercially available today, have bits in micrometer sizes, therefore it would not be possible to reduce them in size significantly using magneto optical techniques, since there is a wavelength limit to which a laser can be focused (a micrometer is already comparable to its wavelength). Some improvements could eventually be made to the writing speed. The current writing speed is around 50 MB/s, which is also comparable to the typical repetition frequency of a laser. But this is not the limit to the speed, knowing that laser pulses can be generated much faster or, eventually, it would be possible to use a cascade of lasers to acquire a speed of writing that exceeds the current speed by a great factor. The other possible improvement would be reducing the contact between the writing head and the magnetic surface since a laser can be focused to a point from a remote location.

7 Conclusion

As the experiments have demonstrated, manipulation of the magnetization with laser pulses is possible in the thermal and non-thermal way. Further experiments are currently being made to investigate the direct effect of light on spontaneous magnetization in other materials and at higher temperatures.

These findings open new insights into the understanding of ultrafast magnetic excitation and, regarding recent progress in the development of compact ultrafast lasers, may provide new prospects for applications of ultrafast photomagnetic phenomena.

Besides, these experiments are also interesting as they help us to get a more detailed picture of the processes in magnetized matter.

8 References


5. Maarten van Kampen, *Ultrafast spin dynamics in ferromagnetic metals*, Proefschrift, 2003, **9, 11**

6. [www.chemweb.org](http://www.chemweb.org), modified by Anton Gradišek

9 Further reading