Optical detection of magnetization: The magneto-optical Kerr effect

I. MAGNETO-OPTICAL PHENOMENA

The linear polarization state of the light is changed when reflecting from the surface, or transmitting through a ferromagnetic material, generally resulting in elliptical polarization. This is demonstrated in Fig. 1. (In the following, we consider the electric component of the electromagnetic field of light to be the polarization vector). The basis of the magneto-optical phenomena is that magnetization in a material breaks time-reversal symmetry, therefore the index of refraction characterizing the material will be different for light beams with right and left circular polarization, $R = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ -i \end{pmatrix}$ and $L = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ i \end{pmatrix}$, being the time-reversed eigenvector pair of the dielectric tensor in the presence of a longitudinal magnetization. Linearly polarized light can be decomposed to the sum of two circularly polarized components with the same magnitude and the opposite sense of rotation. The angle of the polarization with respect to the x-axis, ϕ, manifests as a phase shift between the two circular components of the light, as seen in Eq. 1:

$$\begin{pmatrix} \cos\phi \\ \sin\phi \end{pmatrix} = \frac{1}{2} \begin{pmatrix} 1 \\ i \end{pmatrix} e^{i\phi} + \frac{1}{2} \begin{pmatrix} 1 \\ -i \end{pmatrix} e^{-i\phi}. \tag{1}$$

As the reflection (or transmission) coefficients become different for the two circular polarization states in a magnetic material, the incident linear polarization is changed. For instance, if the index of refraction is purely real (but different) for the two circular components (termed as circular birefringence), an additional phase shift arises between the right and left polarization components, leading to the rotation of the polarization plane of the incident light. Whenever the index of refraction is complex, the incident linear polarization becomes elliptic, with its major axis rotated from the plane of the linear polarization.
The polarization change upon the reflection from a magnetized material is referred to as the magneto-optical Kerr-effect (Fig. 1 a)). This magneto-optical effect is generally very weak, the rotation of polarization plane is typically within the range of $\theta_{Kerr} = 0.001 - 1^{\circ}$. Transmission through an optically transparent magnetic material (Fig. 1 b)) is called the Faraday-effect, which is proportional to the width of the material the light passes through, therefore the effect can be arbitrarily enhanced.

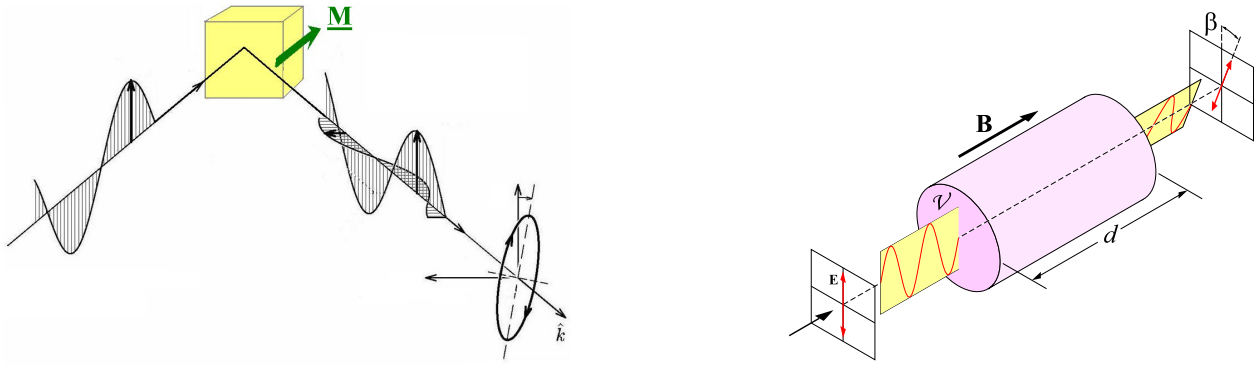

Figure 1: Demonstration of the magneto-optical effects. Left panel: the incident linearly polarized light becomes elliptically polarized reflecting from the surface of a magnetic material. The angle between the major axis of the polarization ellipse and the plane of the incident linear polarization is called Kerr-rotation, while the ratio of the minor and major axes of the ellipse is the Kerr-ellipticity. The change of the polarization can be fully characterized by these two quantities. Right panel: the Faraday-effect, i.e. the rotation of the polarization as the linearly polarized light passes through an optically transparent, magnetized material.

If a material exhibits spontaneous magnetization or is magnetized by an external magnetic field, the time-reversal symmetry of the system no longer holds. In this case, the dielectric tensor characterizing the material with an isotropic or cubic symmetry takes the following form in the presence of finite magnetization along the z axis:

$$\epsilon = \begin{pmatrix} \epsilon_{xx} & \epsilon_{xy} & 0 \\ -\epsilon_{xy} & \epsilon_{xx} & 0 \\ 0 & 0 & \epsilon_{zz} \end{pmatrix} . \tag{2}$$

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Diagonalizing the dielectric matrix for light propagating along the z direction on the subspace of transversal polarization ($\mathbf{E} \in x, y$) yields two circularly polarized eigenstates $R = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ -i \end{pmatrix}$ and $L = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ i \end{pmatrix}$ with the corresponding dielectric eigenvalues ($\varepsilon_{\pm} = \varepsilon_{xx} \pm \varepsilon_{xy}$). The magnitude and phase change of the reflected light is described by the complex-valued reflection coefficient, which is expressed by the Fresnel formula for the reflected light beam with normal incidence:

$$r_{\pm} e^{i\theta_{\pm}} = \frac{1 - n_{\pm}}{1 + n_{\pm}} = \frac{1 - \sqrt{\varepsilon_{\pm}}}{1 + \sqrt{\varepsilon_{\pm}}}. \tag{3}$$

The connection between the Kerr-rotation and Kerr-ellipticity and complex reflection coefficient can be obtained as:

$$\theta_{Kerr} = \frac{\theta_- - \theta_+}{2} \quad \text{and} \quad \eta_{Kerr} = \frac{r_+^2 - r_-^2}{2(r_+^2 + r_-^2)}. \tag{4}$$

Apparently, θ_{Kerr} accounts for the difference in the reflected phase, while η_{Kerr} for the difference in the reflected intensities of the two circular light components. In most of the magnetic materials, the Kerr-parameters are linear functions of the magnetization. Therefore, the measurement of these quantities provide a powerful tool for the optical detection of surface magnetization. Beyond detecting magnetization, by measuring the Kerr-parameters over an extended range of photon energies, i.e. by performing magneto-optical spectroscopy, several fundamental physical properties of the material can be investigated – such as its band structure, crystal-field splittings, the strength of the magnetic exchange and spin-orbit interactions. Recently, besides fundamental research, magneto-optically active materials have been broadly used in optical communication and data storage (e.g. optical isolators, magneto-optical waveguides, magneto-optical data disks, etc.). In this laboratory practice, two novel ferrimagnetic materials with complex crystal structures, $CoCr_2O_4$ and $CuCr_2Se_4$, will be investigated, both exhibiting giant magneto-optical effect.

II. EXPERIMENTAL METHODS

The accurate detection of small changes in the polarization state via the magneto-optical effects requires a sensitive measurement setup. In principle, the magneto-optical Kerr-rotation could be detected by placing the sample between two crossed polarizers (the light is propagating in the direction parallel to the magnetization of the material, with normal incidence to the surface). In this scheme, the light intensity passing through the second polarizer (analyzer) is proportional to the strength of the magneto-optical Kerr-rotation, since in the absence of a Kerr-rotation, no intensity is transmitted through the analyzer. If the analyzer is rotated until minimal (ideally zero) intensity is measured, the Kerr-rotation $\theta_{Kerr}(\mathbf{M})$ could be measured as a function of the sample magnetization, $\mathbf{M}$. The optical path often contains other birefringent elements, such as lenses, causing additional change in the polarization. This baseline can be eliminated if the Kerr-rotation is measured with the reversal of the magnetic field and the Kerr-rotation, being an odd function of the magnetization, is obtained as $\theta_{Kerr} = (\theta(M) - \theta(-M))/2$. However, since the accuracy of setting and reading off the analyzer angle is above 0.1-0.2°, this simple method is not sensitive enough.
A better alternative is to place an additional Faraday-rotator (Fig 1 (b)) between the first polarizer and the sample. By varying the current through the solenoid coil surrounding the Faraday-rotator crystal, the degree of polarization rotation (which is proportional to the magnetic field of the solenoid) can be changed to compensate the magneto-optical rotation of the sample. This method is equivalent with the manual rotation of the analyzer, but a better accuracy can be achieved. For a better understanding, let us follow the change of the polarization as passing through the optical elements shown in Fig. 3.

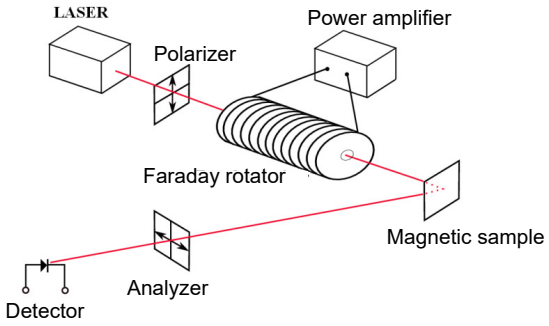

Figure 2: Optical setup for the measurement of the magneto-optical effects: LASER with a wavelength of λ=635nm and 5mW power → vertically aligned polarizer → Faraday-rotator → sample → horizontally aligned analyzer → Si diode photodetector.

We describe the polarization vector in the reference frame of the light. The elements of the polarization vectors are generally complex, representing the temporal phase difference in the polarization components. An ideal Faraday-rotator only rotates the polarization plane, and does not influence the magnitude of the components, as described by the following matrices in the Cartesian and circular base, respectively:

$$F = \begin{pmatrix} \cos\phi_F & -\sin\phi_F \\ \sin\phi_F & \cos\phi_F \end{pmatrix} \leftrightarrow \begin{pmatrix} e^{-i\phi_F} & 0 \\ 0 & e^{i\phi_F} \end{pmatrix}. \tag{5}$$

The reflection matrix of the sample in the circular polarization eigenbase is written as follows:

$$S^{(circ)} = \begin{pmatrix} r_+ e^{i\theta_+} & 0 \\ 0 & r_- e^{i\theta_-} \end{pmatrix}, \tag{6}$$

which needs to be rotated back to the Cartesian reference frame of the light:

$$S^{(Cart)} = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 1 \\ i & -i \end{pmatrix} S^{(circ)} \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & -i \\ 1 & i \end{pmatrix}. \tag{7}$$

Finally, the horizontally aligned analyzer is represented as:

$$A = \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix}. \tag{8}$$

The change of the polarization of the incident vertically polarized light, $\mathbf{E_{in}} = E_0 \begin{pmatrix} 1 \\ 0 \end{pmatrix}$, is obtained as:

$$\mathbf{E} = \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix} \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 1 \\ i & -i \end{pmatrix} \begin{pmatrix} r_+ e^{i\theta_+} & 0 \\ 0 & r_- e^{i\theta_-} \end{pmatrix} \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & -i \\ 1 & i \end{pmatrix} \begin{pmatrix} \cos\phi_F & -\sin\phi_F \\ \sin\phi_F & cos\phi_F \end{pmatrix} E_0 \begin{pmatrix} 1 \\ 0 \end{pmatrix} \tag{9}$$

The photodiode detector, however, measures the intensity of the light, i.e. the temporal average of the absolute value of the electric field strength, squared:

$$I = \frac{E_0^2}{4\pi} \overline{\mathbf{E}^* \mathbf{E}} = \frac{E_0^2}{16\pi} \left[r_+^2 + r_-^2 - 2 r_+ r_- \cos\left(\Delta\theta - 2\phi_F\right) \right], \tag{10}$$

where $\Delta\theta = \theta_+ - \theta- \equiv -2\theta_{Kerr}$. Apparently, the magneto-optical Kerr-rotation of the sample can only be compensated (made zero) by the Faraday-rotator, if $r_+ = r_-$, i.e. the Kerr-ellipticity of the sample is zero, and only Kerr-rotation is present. Otherwise, the light intensity reaching the detector can only be minimized by the Faraday-rotator (or by rotating the analyzer by hand in the previous scheme).
The sensitivity can be further enhanced with the application of high-frequency lock-in detection technique, enabling the elimination of environmental noise originating from other sources of light apart from the laser source, reaching the detector. In order to do so, an ac current with harmonic time dependence is driven through the Faraday-rotator so that: $\phi_F(t) = \phi_F \sin(2\pi f t)$. We exploit that in Eq. 10, the harmonic time dependence in the argument of the cosine function can be expanded in Fourier series in terms of the J_i Bessel's functions:

$$\sin\left[\phi_F \sin(2\pi f t)\right] = 2J_1(\phi_F) \sin(2\pi f t) + \cdots \tag{11}$$

$$\cos\left[\phi_F \sin(2\pi f t)\right] = J_0(\phi_F) + 2J_2(\phi_F) \sin(4\pi f t) + \cdots \tag{12}$$

Furthermore, $r_+ \approx r_-$ and $\theta_{Kerr} \ll 1$, we obtain the following terms in the lowest harmonics in the Fourier expansion:

$$I_f = I_0 \left[4 r_+ r_- \sin(2\theta_{Kerr}) J_1(\phi_F)\right] \approx 4 I_0 J_1(\phi_F) \theta_{Kerr} \tag{13}$$

$$I_{dc} = I_0 \left[r_+^2 + r_-^2 - 2 r_+ r_- J_0(\phi_F) \right] \approx 0, \tag{14}$$

where I_0 is the total intensity measurable with parallel polarizers.

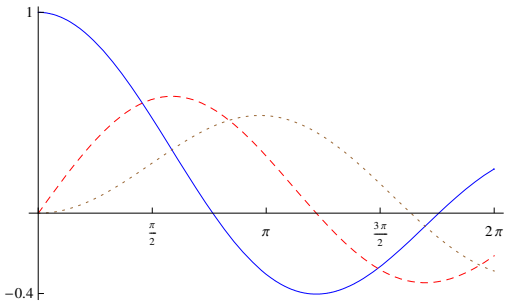

Figure 3: Bessel's functions: $J_0(x)$ blue, $J_1(x)$ red dashed and $J_2(x)$ brown dotted lines.

As seen from Eq. 13, the Kerr-rotation is proportional to I_{1f} modulated intensity. In order to obtain the value of the rotation in degrees, the coefficient $4I_0 J_1(\phi_F)$ must be determined. The most straightforward way to do that is by recognizing that the Kerr-rotation of the sample by an angle of ϕ is equivalent with the rotation of the analyzer by $-\phi$. Thus, the Kerr-rotation can be calibrated by the manual rotation of the analyzer. Furthermore, according to Eq. 13, as the modulation amplitude of the Faraday-rotator is increased, the measured magneto-optical signal becomes larger. Therefore the signal-to-noise ratio of the measurement is expected to improve as the magnitude of the ac current driven through the solenoid in increased. The scheme of the electronic setup is demonstrated in Fig. 4.

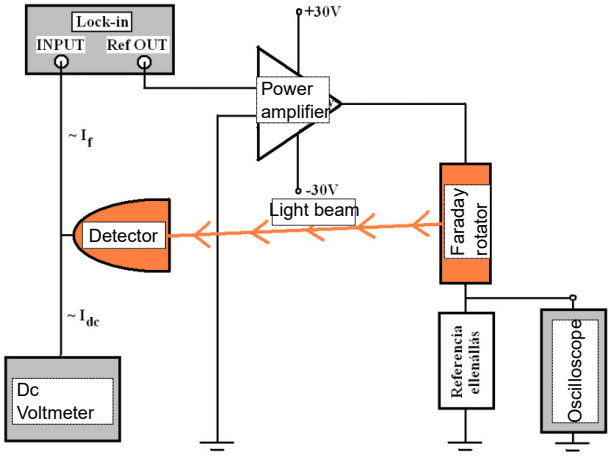

Figure 4: Schematics of the connection of the electronical devices.

III. MEASUREMENT TASKS

We investigate the magnetic properties of two compounds with spinel crystal structure via the measurement of the magneto-optical Kerr effect. The first compound is $CuCr_2Se_4$, which is a room-temperature ferromagnet. The second material is $CoCr_2O_4$, which becomes magnetic only at low temperatures.

- Build the magneto-optical light path and connect the electric devices, using $CuCr_2Se_4$ as the sample. Calibrate the polarization rotation by the manual rotation of the analyzer, determining the prefactor, $4J_1(\phi_F)$.
- Measure the magneto-optical Kerr-rotation of $CuCr_2Se_4$, using a permanent magnet. Determine the magnetic hysteresis of the sample.
- Place the $CoCr_2O_4$ sample located in the low-temperature sample holder into the light path, then use liquid Nitrogen to cool down the sample to $77°C$. Use the solenoid coil fixed to the sample holder to measure the magnetic-field dependence of the Kerr-rotation of the sample at this temperature. Determine the width of the magnetic hysteresis. Repeat this measurement at several temperatures above $77°C$. Determine the temperature of the ferromagnetic phase transition.
- Measure θ_{Kerr} as the function of the temperature, upon the slow heating of the sample in (small) positive and negative magnetic fields.

IV. WARNING!

During measurement, please consider the following hazards:

- The LASER beam has high power and may damage the eyes. Do not look directly into the beam.
- First connect the output of the power amplifier to the solenoid coil and the oscilloscope to the 0.1Ω resistance, BEFORE switching on the power supply of the amplifier. Afterwards, slowly increase the voltage output of the lock-in amplifier, until the measured current (monitored by the oscilloscope connected to the resistance) does not exceed ± 5A.
- Care must be taken when pouring the liquid nitrogen.
- The permanent magnet should be moved carefully. Proximity to magnetic materials may crash the magnet into the object and cause damage to the magnetic or optical elements.

V. FURTHER READING

- 'Bevezetes a modern szilardtestfizikaba', Solyom Jeno (Springer, 2005)
- 'Electronic States and Optical Transitions in Solids', Bassani and Pastori Parravicini (Pergamon, 1975)'
- 'Solid State Spectroscopy' H. Kuzmany (Springer, 1998)
- 'Magneto-optics', S. Sugano and N. Kojima (Springer, 1999)