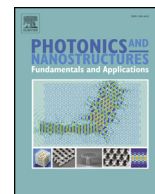




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## Quadratic magneto-optic spectroscopy setup based on photoelastic light modulation



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### ABSTRACT

In most of the cases the magneto-optic Kerr effect (MOKE) techniques rely solely on the effects linear in magnetization ( $\mathbf{M}$ ). Nevertheless, a higher-order term being proportional to  $\mathbf{M}^2$  and called quadratic MOKE (QMOKE) can additionally contribute to experimental data. Handling and understanding the underlying origin of QMOKE could be the key to utilize this effect for investigation of antiferromagnetic materials in the future due to their vanishing first order MOKE contribution. Also, better understanding of QMOKE and hence better understanding of magneto-optic (MO) effects in general is very valuable, as the MO effect is very much employed in research of ferro- and ferrimagnetic materials. Therefore, we present our QMOKE and longitudinal MOKE spectroscopy setup with a spectral range of 0.8–5.5 eV. The setup is based on light modulation through a photoelastic modulator and detection of second-harmonic intensity by a lock-in amplifier. To measure the Kerr ellipticity an achromatic compensator is used within the setup, whereas without it Kerr rotation is measured. The separation of QMOKE spectra directly from the measured data is based on measurements with multiple magnetization directions. So far the QMOKE separation algorithm is developed and tested for but not limited to cubic (001) oriented samples. The QMOKE spectra yielded by our setup arise from two quadratic MO parameters  $G_s$  and  $2G_{44}$ , being elements of quadratic MO tensor  $\mathbf{G}$ , which describes perturbation of the permittivity tensor in the second order in  $\mathbf{M}$ .

### 1. Introduction

The magneto-optic Kerr effect (MOKE) [1] technique is a tool that was and is vastly used for ferro- and ferrimagnetic material research. Setups employing MOKE at a single wavelength are typically used to provide information about magnetic properties of the sample such as magnetic anisotropy, magnetic remanence and coercivity, saturation field, magnetization ( $\mathbf{M}$ ) reversal process or detection of exchange bias. On the other hand, setups providing MOKE response over a continuous spectrum (usually extended visible spectral range) yield the information about the electronic structure of the sample (here the results are usually accompanied by ab-initio calculations) [2–13]. Note that in both cases, one is usually relying on MOKE linear in  $\mathbf{M}$  (LinMOKE). MOKE quadratic in  $\mathbf{M}$ , quadratic MOKE (QMOKE), did not get large attention yet and is often considered as small parasitic effect. Nevertheless, the QMOKE effects are often accompanying LinMOKE

measurements, and hence its clear understanding is important. Further, the fact that QMOKE is even in  $\mathbf{M}$  makes it applicable for investigating antiferromagnetic materials which do not have a LinMOKE response [14,15].

At the beginning of the 90s unexpected symmetric contributions to the hysteresis loops of Ni-Fe bilayers were reported [16,17] and later on explained as QMOKE contributions to the overall MOKE signal [18–20]. Several methods have been proposed for the separation of QMOKE contributions from the LinMOKE signal including the ROTMOKE method [21], the 8-directional method [22], the sample rotation by 180° [23], and the rotation field method [24]. Here we present QMOKE spectroscopy setup, which is capable to measure two types of QMOKE spectra in the spectral range of a 0.8–5.5 eV, QMOKE  $\sim G_s$  spectra and QMOKE  $\sim 2G_{44}$ , where  $G_s$  and  $2G_{44}$  are quadratic magneto-optic (MO) parameters that fully describe the perturbation to the permittivity tensor in the second order in  $\mathbf{M}$  of materials with cubic crystallographic

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structure [25–28]. LinMOKE spectra, namely longitudinal MOKE (LMOKE) spectra, can be measured as well.

The measurement process is based on light modulation using photoelastic modulator and consequent detection by a lock-in amplifier. For the separation of LMOKE and of each QMOKE contribution, we employed a technique similar to the 8-directional method, but using a combination of just 4 directions and a sample rotation by 45° as described in this article.

## 2. Theory of MOKE

### 2.1. Description of polarized light and MOKE

MOKE manifests through the change of the polarization state of light reflected from a magnetized sample. Generally elliptically (fully) polarized light can be described by the Jones formalism using Jones vector such as [29]

$$\mathbf{J}_{\theta\epsilon} = \begin{bmatrix} \cos \theta \cos \epsilon - i \sin \theta \sin \epsilon \\ \sin \theta \cos \epsilon + i \cos \theta \sin \epsilon \end{bmatrix}, \quad (1)$$

where  $\theta$  is the rotation angle of major axis of the polarization ellipse in our coordination system and  $\epsilon$  is the ellipticity, i.e. arctan of the ratio of the minor and major axis of the polarization ellipse. Through those two values  $\theta$  and  $\epsilon$ , an arbitrary state of full polarization can be described. The complex polarization parameter  $\Phi$  is then defined as the ratio of the second and first component of the Jones vector  $\mathbf{J}_{\theta\epsilon}$

$$\Phi = \frac{\sin \theta \cos \epsilon + i \cos \theta \sin \epsilon}{\cos \theta \cos \epsilon - i \sin \theta \sin \epsilon} = \frac{\tan \theta + i \tan \epsilon}{1 - i \tan \theta \tan \epsilon}. \quad (2)$$

In case of small angle approximation ( $\theta < 1^\circ$ ,  $\epsilon < 1^\circ$ ) we can write

$$\Phi = \theta + i\epsilon. \quad (3)$$

Within Jones formalism, the reflection of polarized light from a sample is described by the reflection matrix [29,30]

$$\mathbf{R} = \begin{bmatrix} r_{ss} & r_{sp} \\ r_{ps} & r_{pp} \end{bmatrix}, \quad (4)$$

where the first lower index of the elements refers to the polarization state of reflected light, while the latter index to the polarization state of the incident light. For isotropic systems without magnetization  $r_{sp} = r_{ps} = 0$ . If the sample gets magnetized, a certain part of the reflected  $s$ -polarized wave is converted into a  $p$ -polarized wave (or vice versa). This change of polarization is described by the complex Kerr amplitude  $\Phi_{s/p}$  for  $s$  and  $p$  polarized incident light.  $\Phi_{s/p}$  is actually analogous to the complex polarization parameter  $\Phi$  from Eq. (3). With respect to the Jones formalism and Eq. (4) [29,31]

$$\begin{aligned} \Phi_s &= \theta_s + i\epsilon_s = -\frac{r_{ps}}{r_{ss}}, \\ \Phi_p &= \theta_p + i\epsilon_p = \frac{r_{sp}}{r_{pp}}. \end{aligned} \quad (5)$$

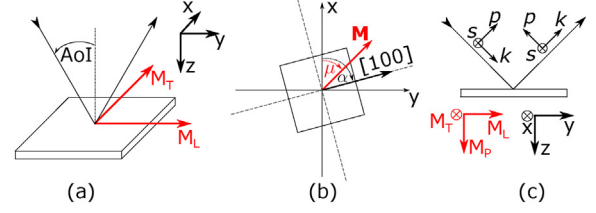
Here  $\theta_{s/p}$  is called Kerr rotation and  $\epsilon_{s/p}$  is called Kerr ellipticity and in common called Kerr angles.

### 2.2. Theory of linear and quadratic MOKE

The reflection coefficients from Eq. (5) are strictly bound with the permittivity tensor  $\epsilon$  of the crystal. The permittivity tensor elements of ferromagnetic (FM) material,

$$\epsilon_{ij} = \epsilon_{ij}^{(0)} + K_{ijk}M_k + G_{ijkl}M_kM_l, \quad (6)$$

are described up to second order in magnetization  $\mathbf{M}$  by the permittivity in the 0th order in  $\mathbf{M}$  ( $\epsilon^{(0)}$ ) and by the linear and quadratic MO tensor  $\mathbf{K}$  and  $\mathbf{G}$ , respectively [32].  $M_k$  and  $M_l$  are components of normalized  $\mathbf{M}$ . In case of the cubic crystal structure (classes  $\bar{4}3m$ ,  $432$ ,  $m\bar{3}m$ ), the linear MO tensor is described by one free parameter  $K$  and the quadratic MO tensor is described by two free parameters  $2G_{44}$  and  $G_s = (G_{11} - G_{12})$



**Fig. 1.** (a) Right-handed coordinate system  $\hat{x}$ ,  $\hat{y}$ ,  $\hat{z}$  is established with respect to the plane of incidence and surface of the sample. Components of the in-plane normalized  $\mathbf{M}$ ,  $M_T$ ,  $M_L$  are defined along axis  $\hat{x}$ ,  $\hat{y}$  of the coordinate system, respectively. The AoI stands for angle of incidence. (b) Definition of positive in-plane rotation of the sample and  $\mathbf{M}$  within the  $\hat{x}$ ,  $\hat{y}$ ,  $\hat{z}$  coordinate system, described by angle  $\alpha$  and  $\mu$ , respectively. (c) Definition of the right-handed Cartesian system  $\hat{s}$ ,  $\hat{p}$ ,  $\hat{k}$  of incident and reflected beam. All directions and angles shown in the figure are of positive values.

[28,31,32]. The permittivity tensor in the 0th order in  $\mathbf{M}$  is described by the scalar  $\epsilon_{(d)}$ ,  $\epsilon^{(0)} = \epsilon_{(d)}\mathbf{I}$ , where  $\mathbf{I}$  is identity matrix.

The analytical approximation of MOKE response for FM layers is [25,33]

$$\begin{aligned} \Phi_s &= -\frac{r_{ps}}{r_{ss}} = A_s \left( \epsilon_{yx} - \frac{\epsilon_{yz}\epsilon_{zx}}{\epsilon_{(d)}} \right) + B_s \epsilon_{zx}, \\ \Phi_p &= \frac{r_{sp}}{r_{pp}} = -A_p \left( \epsilon_{xy} - \frac{\epsilon_{zy}\epsilon_{xz}}{\epsilon_{(d)}} \right) + B_p \epsilon_{xz}, \end{aligned} \quad (7)$$

where  $A_{s/p}$  and  $B_{s/p}$  are the optical weighting factors that are even and odd functions of the angle of incidence (AoI), respectively (and in the latter case we can approximately write  $B_{s/p} \sim \sin(\text{AoI})$ ). Further they are also functions of optical surroundings of FM layer, i.e. substrate, overlayer and other layers in the stack.

If we will limit ourselves to in-plane normalized  $\mathbf{M}$  and cubic crystal structure with [001] direction normal to the surface, then the dependence of the MOKE amplitude on the MO parameters  $K$ ,  $G_s$ ,  $2G_{44}$ , on the sample orientation  $\alpha$  and on the in-plane  $\mathbf{M}$  direction  $\mu$  (see Fig. 1) can be derived from Eqs. (6) and (7) [25,27,34], resulting in

$$\begin{aligned} \Phi_{s/p} &= \pm A_{s/p} \left\{ \frac{2G_{44}}{4} [(1 + \cos 4\alpha) \sin 2\mu + \sin 4\alpha \cos 2\mu] \right. \\ &\quad \left. + \frac{G_s}{4} [(1 - \cos 4\alpha) \sin 2\mu - \sin 4\alpha \cos 2\mu] \right\} \\ &\quad \mp A_{s/p} \frac{K^2}{2\epsilon_{(d)}} \sin 2\mu \\ &\quad \pm B_{s/p} K \sin \mu. \end{aligned} \quad (8)$$

The sign  $\pm$  is given by the polarization ( $s/p$ ) of the incident light beam. This dependence provides us with measurement sequences, which isolate the individual MOKE contributions that have its origin in the individual MO parameters [22]. These measurement sequences with exact definitions of the angles  $\alpha$  and  $\mu$  will be discussed in Sections 3.1 and 3.3.

## 3. Setup description

### 3.1. Conventions and definitions

At first, we have to introduce conventions for positive and negative rotations of crystallographic structure, of  $\mathbf{M}$ , of light polarization and of optical elements. We further have to define all coordinate systems used within the setup in which those rotations take place. To describe reflection on the sample, three Cartesian systems are needed, one for the incident light beam, one for the reflected light beam and one for the sample.

- (i) The electric field vector of an electromagnetic wave is described by a negative time convention as  $\mathbf{E}(\mathbf{r}, t) = \mathbf{E}(\mathbf{r})e^{-i\omega t}$ , providing permittivity in form of  $\epsilon = \epsilon_R + i\epsilon_I$ , where  $\epsilon_R$ ,  $\epsilon_I$  being real, imaginary part of complex permittivity, respectively, where  $\epsilon_I > 0$ .

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