Optical Materials 55 (2016) 115–120

Contents lists available at ScienceDirect

Optical Materials

journal homepage: www.elsevier.com/locate/optmat

Magnetooptical spectrometer based on photoelastic modulator with optical feedback and its application in study of *f*-electron materials

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ARTICLE INFO

Article history:
Received 30 January 2016
Received in revised form 24 February 2016
Accepted 7 March 2016
Available online 21 March 2016

Keywords:
Photoelastic modulation
Magnetic circular polarized luminescence
Faraday rotation
Rare-earth garnets

ABSTRACT

A spectroscopic apparatus for measurement of magnetooptical and optical properties of *f*-electron materials has been designed and established using a polarization modulation technique based on photoelastic modulator with optical feedback. The magnetooptical system is able to provide a sensitivity of 0.004 arc-deg at the wavelength of 380 nm. The versatile applications of the spectrometer have been verified by the measurements of the magnetic circular polarized luminescence and Faraday rotation angle in holmium- and terbium-containing garnet crystals.

1. Introduction

Magnetooptical spectroscopy is an advanced and important technique in characterization of solid state materials. Magnetooptics gives not only information on influence of magnetic field on energy levels of a quantum system (Zeeman polarization spectroscopy), but also information on energy structure of both 3*d* transition-metal ions and 4*f* rare-earth ions in compound crystals. It may also give physical insights into interband and intraband optical transitions in semiconductors and metals, revealing symmetry of paramagnetic centers and displaying magnetic domain patterns.

At present time to obtain specific information on excited states of 3*d*- and 4*f*-ions, magnetic circular polarized luminescence (MCPL) technique is frequently utilized. The "absorbing" analog of MCPL is the widely used method of magnetic circular dichroism (MCD). Both techniques allow measuring of important physical parameters such as Zeeman splitting, *g*-factors, ratio between velocities of the radiative and spin-lattice relaxations in the excited state, and degree of thermalization of the excited states.

For realization of the dedicated magnetooptical measurement, it is necessary to use polarization modulation techniques, among them the method of elliptical polarization modulation is commonly applied [1,2]. This technique operates on the basis of different polarization modulators, such as Faraday rotators, Pockels cells, and photoelastic modulators [3,4]. In practice photoelastic modulation has shown special advantages and is frequently utilized. The principles of polarization modulation using photoelastic modulators have been given in Refs. [5,6]. Modulation of the light ellipticity caused by oscillation in the bar of an isotropic material is employed in the optical spectroscopy and ellipsometry [5,6]. The main advantage is its capability to measure signals with ~100% depth of modulation, which is very important in the case of strong absorption and weak reflection or emission. In addition, for such kinds of modulators broad-aperture beams can be used [5].

In the present study we have designed and established a magnetooptical spectrometer based on the light modulation created by the photoelastic modulator of an original design [8]. The system has been tested by measurements of the MCPL degree and Faraday rotation (FR) angle in the holmium- and terbium-containing garnet crystals, carried out in strong magnetic field using different light sources.

2. Basic principles of measurement of the linear magnetooptical effects

It is well-known that the mechanical longitudinal oscillations in the quartz bar lead to the periodical modulation of its index of refraction along the largest dimension due to the photoelastic

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http://dx.doi.org/10.1016/j.optmat.2016.03.011

effect [5,6]. If the light wave propagates in the quartz bar perpendicularly to the mechanical oscillations the phase shift φ arising between orthogonal components of the electrical vector of the light wave caused by the induced birefringence can be written as:

$$\varphi = \frac{2\pi\Delta n}{\lambda} \sin\Omega t \cdot l \tag{1}$$

where Δn is the maximal difference of the indexes of refraction of two orthogonal light components passing through the active element of the modulator, Δn is a function of the maximal deformation in the quartz bar, which depends on the voltage supply of the active element of the modulator, λ is the light wavelength in vacuum, l is the thickness of the quartz bar along the light propagation, and Ω is the operation frequency. If the incident light flux of the linear polarized light upon the modulator has non-zero components along the orthogonal axes and if one of them is parallel to the direction of the deformation of the modulator active element, then the polarization state of the light passing through the bar becomes modulated.

2.1. Measurement of MCPL and MCD

In our magnetooptical measurements using passing (MCD) or emitted (MCPL) light, so-called natural (completely non-polarized) light radiation is projected onto the sample under study, which is located in a magnetic field. The orientation of the magnetic field is parallel to the light propagation. The light outgoing the sample in the magnetic field is partially-polarized. In given geometry of the experiment the light is partially *circularly*-polarized and can be characterized by the degree of the circular polarization P:

$$P = \frac{\Phi_+ - \Phi_-}{\Phi_+ + \Phi_-} \tag{2}$$

where Φ_+ and Φ_- are fluxes of the right- (+) and left- (−) circular polarized incoherent light components. The degree of the circular polarization P can be measured with a photoelastic modulator and a linear polarizer [5].

Light intensity outgoing the photoelastic modulator has constant and alternative components and can be represented as series of harmonics:

$$\begin{aligned}\Phi &= \frac{\Phi_+ + \Phi_-}{2} + \frac{\Phi_+ - \Phi_-}{2} \sin\varphi \\ &= \frac{\Phi_+ + \Phi_-}{2} + \frac{\Phi_+ - \Phi_-}{2} [2J_1(\varphi_M) \sin\Omega t + \cdots] \\ &= \Phi(0) + \Phi(\Omega) + \cdots \end{aligned} \tag{3}$$

where φ is the phase shift caused by the birefringence given by (1), $J_i(\varphi_M)$ are Bessel functions and φ_M is the maximum phase shift induced by the modulator at a certain voltage supply. In the series of harmonics in (3) the Bessel functions with indexes $i > 1$ can be experimentally removed using lock-in amplifier tuned in on signal of frequency Ω, i.e., all multiple to Ω frequencies are filtered by the lock-in, then (2) can be rewritten as:

$$\frac{I(\Omega)}{I(0)} = \frac{K_1\Phi(\Omega)}{K_2\Phi(0)} = 2J_1(\varphi_M)KP \sin\Omega t = AP \sin\Omega t \tag{4}$$

where $K_{1,2}$ are the coefficients of proportionality between corresponding currents of the photomultiplier tube (PMT) and the intensities of the light; $I(\Omega)$ and $I(0)$ are the corresponding alternative and constant PMT currents; $A = 2J_1(\varphi_M)K$ is an instrument constant at a certain wavelength; $J_1(\varphi_M)$ and therefore $I(\Omega)$ are maximal at $\varphi_M = 105$ deg. Practically the constant A is found at the calibration procedure using circular polarizer ($P = 1$) instead of the sample. Thus the ratio of two measured signals $I(\Omega)/I(0)$ determines the magnitude of the degree P of the partially circularly-polarized light.

On the one hand, the degree of the partially circularly-polarized light P measured in the study of MCPL spectra defines the relative difference of the contributions of the orthogonal circularly-polarized emission transitions induced by the external magnetic field. On the other hand, the partially circularly-polarized degree measured in the MCD spectra induced by the magnetic field defines the difference of the orthogonal circularly-polarized absorption coefficients of light, i.e., $P = ½(\alpha_+ - \alpha_-)l$, where $\alpha_\pm$ are the absorption coefficients for the orthogonal circularly-polarized light components and l is the thickness of the sample.

In present study, we use modulation of light polarization generated by the photoelastic modulator [7]. An active element of the modulator is a bar of isotropic fused quartz with dimensions of $75 \times 20 \times 12$ mm^3 which is connected with a rectangular piezo-ceramic transducer. Both parts of the modulator operate at the same resonance frequency of 36 kHz. The voltage supply of the modulator is 30–90 V depending on the light wavelength under investigation. For voltage supplying of the photoelastic modulator an active oscillator scheme is used, shown in Fig. 1. The output of the scheme is connected with a piezo-ceramic transducer. As a basis for the realization of the positive feedback we use an optoelectronic couple mounted on the modulator. To control the oscillation phase the sinusoidal signal from the optoelectronic couple is fed to the input of the active oscillator scheme, in detail considered in Refs. [6,8]. Application of such electrically isolated optical positive feedback gives rise to increased reliability at starting and operation of the modulator [8].

Fig. 1 shows a schematic diagram of the magnetooptical device for measuring MCPL. The light radiation of a 500 W Xe-lamp is collected by a quartz condenser, passes through a water- and UV-filters with a bandpass of 250–400 nm, then is focused on the sample located in the air-gap of the magnet, and finally excites a photoluminescence. In the magnetic field H the photoluminescence emitted by the sample is partially-polarized. Next, the partially-polarized light collected by the objective lens, passes through a photoelastic modulator and a linear polarizer acting as analyzer, and is focused on the input slit of the monochromator (Model HISW50). The induced optical axis of the photoelastic modulator and the transmission plane of the analyzer are rotated on 45 deg relative to each other. The photomultiplier (Model FEU-79, 71 or 100) operates in the constant average current mode with using of the adjustable voltage regulator UA723C with negative feedback in the photomultiplier anode circuit. Note that the stabilizer provides anode current with a stabilization factor of 1–2%. It is significant that in the constant average current mode, the measured value of the ratio $I(\Omega)/I(0)$ is simply proportional to the measured signal $I(\Omega)$, since the denominator of the ratio (4) is constant regardless of the illumination change of $\Phi(0)$ on the PMT photocathode [9].

It is important to note that in the MCD spectra measuring the experimental device is completely identical to that used for measurement of the MCPL degree spectra, but here a halogen lamp is used instead of the Xe-lamp, which is more convenient to perform magnetooptical studies in the visible region.

Moreover, the designed setup also provides measurement of the optical absorption spectra using the same optical configuration. Indeed, in realization of the PMT average current stabilization (i.e., in the mode of $\bar{I}_P = I_p = const$) due to the feedback between dc-amplifier connected with the output of the PMT and its high voltage source, one can obtain ratio between the relative change $\Delta\Phi/\Phi$ of light flux Φ passing the sample and PMT voltage supply U of the PMT. In a linear region of the current-illumination characteristic of the PMT there is ratio between anode current I_P, light flux Φ, and voltage supply U:

$$I_P = A \cdot \Phi \cdot f(U) \tag{5}$$