OPTICAL REFLECTION AND POLAR KERR ROTATION OF COBALT AND TITANIUM SUBSTITUTED BARIUM HEXAFERRITES

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Abstract - Optical reflection and the polar Kerr rotation of four single crystals of barium hexagonal ferrites were measured in the visible and near infrared region at room temperature. Local extrema of the polar Kerr rotation at wavelengths of 1650, 1350 and 760 nm could be assigned to the presence of Co^{2+} in tetrahedral positions. The figure of merit attains the maximum values at these wavelengths suggesting material to be suitable for the magneto-optical storage applications. KEYWORDS: HEXAGONAL FERRITES, MAGNETO-OPTICAL PROPERTIES, FIGURE OF MERIT.

INTRODUCTION

Cobalt and titanium substituted barium hexaferrite (CoTiBaM) powders and thin films proved to be an excellent material for the high density magnetic storage of information. In order to achieve a better tailoring of magnetic properties of fine particles and/or thin films, the properties of bulk material of the same chemical composition must be well understood. Only little optical and magneto-optical data on single crystal samples were available until recently [1]. In our preceding papers the magnetic, optical and the magneto-optical characteristics of the flux-grown single crystals of CoTiBaM were reported [2-5]. Although the crystals were the best ones available at that time, the flux growth inevitably introduced some concentration gradients and imbedded certain amount of foreign atoms into the hexagonal lattice. Measurements of the optical reflection and the polar Kerr rotation on the natural cleaved surfaces [2] with a number of the growth steps might have induced the interference effects that led to the unusually high and irregular spectral and angle dependences of the PKR on these flux-grown crystals. Consequently, new reliable optical and magneto-optical data for these materials are desirable.

In the present paper the spectral dependences of the optical reflection (OR) and the polar Kerr rotation (PKR) of four high-quality single crystals of hexagonal ferrites are studied in order to get information about the spectral regions possessing the best figure of merit. The quality of the present samples that were prepared by the floating zone-melting and additionally heat-treated in the controlled atmospheres is much better than that of the flux grown crystals used in the past [1-5]. Besides the commonly studied Co, Ti hexaferrite series also a new BaTiFe_{12}O_{19} single crystal was synthesized to find out the influence of the two-valent iron ions on the OR and PKR spectra.

EXPERIMENT

Single crystals of BaCo_{x}Ti_{1-y}Fe_{12-x-y}O_{19} hexagonal ferrites, i.e. BaM (x=y=0), BaTi (x=0, y=1) and CoTiBaM (x=y=0.2 and 0.8) were prepared by the arc-image floating zone-melting of the corresponding polycrystalline rods as described earlier [6]. After the growth process single crystal cylinders of about 5 mm in diameter and several cm in length were additionally heat-treated and cooled in controlled atmospheres to remove mechanical stresses and to improve the oxygen stoichiometry and the chemical homogeneity. From the single crystal bars the (0001) oriented slices were cleaved and some of them ground to 100 μm thick platelets. Upper faces were polished to mirror finish to enable reliable OR and PKR spectra measurements free of the influence of the cleavage steps.

Spectral dependences of the optical reflectances were measured in the wavelength range from 500 to 1600 nm at room temperature by an absolute method with the use of the integration sphere to take account of the nonspecular reflections. Polar Kerr rotations in the 400 to 2000 nm wavelength region were measured with the light propagating almost perpendicularly to the (0001) hexagonal planes. The only exception was the BaM sample in which the polished surface contained (0001) axis. External magnetic fields of 0.5 tesla were applied perpendicularly to the polished surfaces, i.e. along the direction of light propagation. The applied fields were sufficient to saturate all samples except of BaM where the correction for a nonsaturated state has been made. Polarization modulation method as described earlier [2,7] was utilized to measure PKR in the whole investigated region.

RESULTS

Spectral dependences of the optical reflectivity as
a function of the photon energies of the incident light are plotted in Fig. 1 for samples without cobalt and in Fig. 2 for cobalt containing samples. Good agreement within 2-3% is found with our previous results obtained on the polished surfaces of the flux-grown samples [3].

Spectral dependences of the polar Kerr rotation of all samples studied are represented in Fig. 3. There are no significant spectral features in PKR of BaM in agreement with the early papers [1,2]. Substitution of titanium (i.e. BaTi sample) modifies the almost constant Kerr rotation values of BaM into the spectral dependent curve that reminds PKR behaviour of magnetite [7-9]. Also, the introduction of cobalt induces new spectral features that are complementary to those observed in the Faraday rotation spectra of Co-Ti-substituted hexaferrite platelets [3] or thin films [10-12].

In contrast to the unusually large PKR values (about 1 degree) obtained on the flux grown single crystals earlier [2] the present local extrema observed at photon energies of about 0.7, 0.9 and 1.7 eV (i.e. at wavelengths of 1650, 1350 and 760 nm, respectively) attain much smaller values (less than 0.1 degree). This supports our early suspicion about the former unrealistically large PKR values [2] that are obviously due to the light interference on the growth steps of cleavage surfaces. Consequently, large PKR values reported in [2] do not represent the inherent properties of these kinds of material. The present OR and PKR measurements thus correct the previous results.

**DISCUSSION**

To assess suitability of the material for the magneto-optical applications the spectral dependence of the figure of merit has to be evaluated. There is no unique definition of figure of merit for the reflection arrangement of the experiment [13,14]. The most suited for the optical reflectivity (OR) and the polar Kerr rotation (PKR) measurements is the definition by [14] which relates the absolute value of PKR and the square
Materials attaining the maximum values of $m$ range among the best for the applications. In Fig. 4 spectral dependences of $m$ for all the samples studied are demonstrated. The two infrared extrema (for the photon energies less than 1 eV) have only a minor significance but the maximum at 1.76 eV (i.e. at 700 nm) falls into the important light region of the GaAs semiconductor lasers. The peaking values of $m$ for CoTiBaM sample $x=y=0.8$ are fairly large suggesting that the material can be used for the magneto-optical storage and integrated optics applications.

![Figure 4](image)

**Fig. 4.** Spectral dependence of the figure of merit, $m$.

Spectral dependences of $m$ also clearly demonstrate various ions contributions to magneto-optical activities of the materials. Broad and structureless behaviour of BaM and BaTi samples points to the collective charge transfer transitions due to the iron ions [9]. The narrow, well localized and concentration dependent peaks of $m$ in the cobalt containing hexaferrites, on the other hand, indicate electron transitions among the localized crystal field split 3d-levels of cobalt ions.

In agreement with the results of the Faraday rotation measurements [3,4,12] we conclude that the magnitudes of the peaks depend linearly on the cobalt concentration. In particular, sharp peaks in the infrared (0.75 and 0.9 eV) are assignet to $^4A_2 \rightarrow ^2T_1(^F)$ type transitions and the peak at the visible red light (1.76 eV) is due to $^4A_2 \rightarrow ^2T_1(^P)$ electron transitions of Co$^{2+}$ in tetrahedral positions of hexagonal lattice. Appropriate substitution of cobalt ions thus can fine tune desired properties of hexaferrites.

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**References**

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