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# Magneto-Optical Properties of Polycrystalline $CoCrFeO_4$ Films

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**Abstract**—A methods of obtaining polycrystalline $CoCrFeO_4$ films based on solid-phase reactions in $Cr_2O_3$/Co/Fe layer structures in the regimes of isothermal annealing and self-propagating high-temperature synthesis (SHS) is described. The magneto-optical properties of the obtained films have been studied. It is established that the spectra of magneto-optical characteristics depend on the solid-phase reaction regime and synthesis temperature. The Faraday rotation angle exhibits a local maximum ($2\theta_F = 5$ deg/μm) at a wavelength of 630 nm.

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In recent years, considerable effort has been devoted to investigations of the magnetophotonic crystals (MPCs) that represent a new class of heterogeneous materials [1]. The interest in these crystals is related to both the applied aspects and new physical phenomena observed in these media. One-dimensional MPCs typically appear as multilayer structures comprising a magnetic layer sandwiched between identical multilayer periodic structures of dielectrics possessing various optical properties. In some MPCs, the magnetic layer represents a ferrimagnetic film, in particular, garnet ferrite film [2]. Epitaxial films of Bi-substituted garnet ferrite for many years demonstrated record high magneto-optical characteristics [3]. As is known, ionic compounds of cobalt, including oxide spinels, exhibit high values of the Faraday rotation angle in the visible spectral range [4, 5]. In this context, it was of interest to synthesize polycrystalline films of magnetic oxides with high values of magneto-optical characteristics. The task of creating MPCs imposes certain conditions on the methods that can be used to obtain both magnetic and dielectric layers. The most acceptable approach is provided by the vacuum deposition techniques.

This Letter presents the results of investigations of the magnetic and magneto-optical properties of polycrystalline $CoCrFeO_4$ ferrite films synthesized using solid-phase reactions in metal/oxide layer structures at relatively low temperatures. The solid-phase reactions can proceed in the regimes of either isothermal annealing or self-propagating high-temperature synthesis (SHS) [6–8]. The SHS is possible if the initial mixture contains a fuel and an oxidizer, which are necessary for the reaction to proceed in the regime of combustion. For the synthesis of oxides, the fuel is typically a metal, the role of an oxidizer is played by oxygen, and the main reaction is metal oxidation, which ensures the evolution of heat necessary for the SHS. Oxygen can be supplied from both internal and external sources.

We have obtained cobalt ferrite-chromite films using the following solid-phase reaction:

$$Co + Fe + 0.5Cr_2O_3 + 1.25O_2 = CoCrFeO_4. \quad (1)$$

Reactants were represented by layers in a $Cr_2O_3$/Co/Fe film structure, which were deposited onto fused silica substrates by the sequential evaporation of metals in vacuum ($5 \times 10^{-6}$ Torr) at a substrate temperature of 470 K. Prior to the deposition of Co and Fe, the Cr layer (deposited onto the substrate in the preceding step) was oxidized in air.

The solid-phase reaction in thin-film structures were carried out in regimes of either isothermal annealing (at 750 and 900–950 K) or SHS in air. For the ferrite synthesis in the SHS regime, a film structure was placed onto a tungsten heater and heated at a rate of no less than 20 K/s (thermal explosion) until reaching the SHS wave initiation temperature (~900 K). Then, the combustion front propagation could be observed visually and was typical of the SHS of thin films [6].

The solid-phase reaction yielded polycrystalline cobalt ferrite-chromite films with thicknesses within 150–200 nm. The chemical composition and thickness of each film were determined using X-ray fluorescence (XRF) measurements. The crystalline structure was studied by X-ray diffraction (XRD). The perpendicular magnetic anisotropy constant and saturation magnetization were measured on a torsion magnetometer. The hysteresis loops of the polar Kerr effect and the spectral dependence of the Faraday rotation were measured using a monochromator ($\lambda$ = 400–900 nm) in a magnetic field of up to 16 kOe.

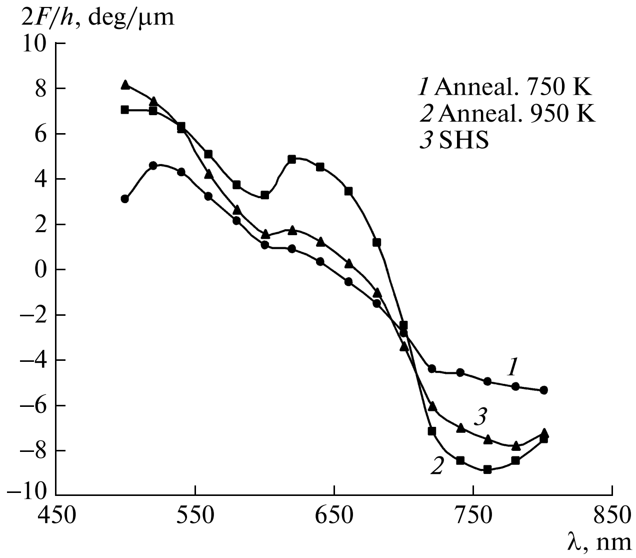


**Fig. 1.** Spectral dependences of the Faraday rotation angle for polycrystalline $CoCrFeO_4$ films synthesized in various regimes.

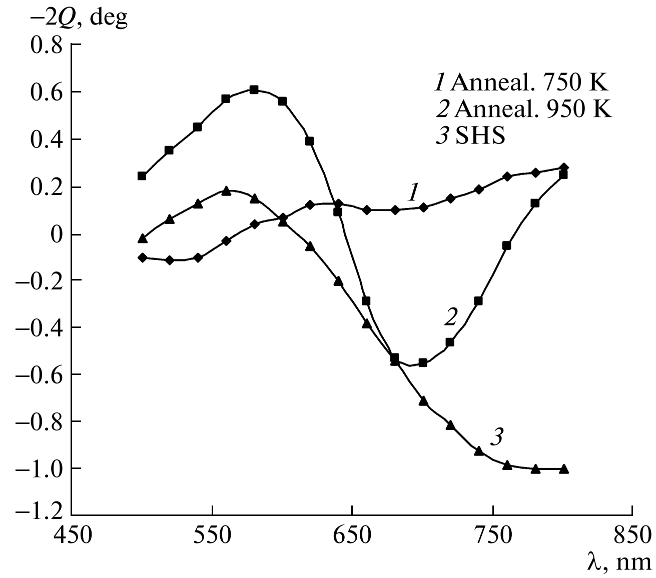


**Fig. 2.** Spectral dependences of the Kerr rotation angle for polycrystalline $CoCrFeO_4$ films synthesized in various regimes.

The XRD patterns of ferrite films obtained by solid-phase reactions in $Fe_2O_3$/Co/Cr layer structures displayed only reflections of a spinel ferrite, irrespective of the regime.

The curves of magnetization reversal in polycrystalline cobalt ferrite-chromite films were obtained by measuring the field dependence of the optical rotation angle in the polar Kerr effect at $\lambda = 800$ nm. It was established that the ferrite films obtained using SHS and the isothermal annealing at 950 K are characterized by higher values of the rectangularity coefficient ($S$) and perpendicular magnetic anisotropy constant ($K_\perp$): $S = 2\theta_K(H_0)/2\theta_K(H_{max}) = 0.9–0.95$, $K_\perp = (5–7) \times 10^5$ erg/cm$^3$. The coercive field of these films was within $H_c = 2–3$ kOe and the saturation magnetization amounted to $M_s = 200$ G.

Figure 1 shows the spectral dependences of the Faraday rotation angle for cobalt ferrite-chromite films synthesized by isothermal annealing at different temperatures and in the SHS regime. Note a positive peak appearing at $\lambda = 630$ nm, which is characteristic of cobalt ferrite-chromite [4]. The value of $2\theta_F = 5$ deg/μm at $\lambda = 630$ nm, which was observed for the films synthesized at 900–950 K, is several times greater than the corresponding values reported for cobalt ferrite films with the same concentration of $Co^{2+}$ ions [9]. The Faraday rotation at $\lambda = 800$ nm is also rather high and exceeds the level known in other ferrites (spinels, garnet ferrites).

The observed increase in Faraday rotation for cobalt ferrite-chromite at a wavelength on the He–Ne laser can be explained as follows. The magneto-optic spectra of spinel ferrites in the visible and IR spectral range are related to the *d*–*d* transitions in magnetic ions occupying octahedral and tetrahedral positions. In cobalt ferrite, $Co^{2+}$ ions in tetrahedral positions mostly contribute to these spectra at $\lambda = 630$ nm. It is also known that chromium ions preferentially occupy octahedral positions in the spinel structure. As a result, $Co^{2+}$ ions occur in tetrahedral positions, which leads to the enhancement of magneto-optic rotation at the indicated wavelength for cobalt ferrite when a fraction of $Fe^{3+}$ ions is replaced by $Cr^{3+}$ ions. An equilibrium state of this ferrite with a spinel structure corresponds to the normal spinel, whereby divalent ions are completely arranged in the tetrahedral positions. The dependence of the Faraday rotation spectrum on the regime of solid-phase reaction is probably related to a difference in the reaction rate under isothermal annealing and SHS conditions. The process of film formation in the regime of annealing is more equilibrium than the SHS, which in fact represents a thermal explosion. Therefore, the films obtained using SHS can possess a partly inverse spinel structure, which leads to a decrease contribution of "tetrahedral" $Co^{2+}$ ions to the magneto-optic spectrum at $\lambda = 630$ nm. Note also that the character of the magneto-optic spectra of the obtained ferrite films confirms the presence of cobalt in the tetrahedral positions of the spinel structure [4] and, hence, the effect of $Cr^{3+}$ ions on the distribution of $Co^{2+}$ ions between the crystal sublattices.

Figure 2 presents the spectral dependences of the Kerr rotation angle for cobalt ferrite-chromite films synthesized by isothermal annealing at different temperatures and in the SHS regime. As can be seen, the spectrum of a film synthesized at 750 K shows almost no evidence of resonances characteristic of cobalt ferrite. This fact can be related to an imperfect crystalline structure formed at the relatively low temperature. The character of the magneto-optic spectrum of the film synthesized at 950 K corresponds to the known spectrum of cobalt ferrite-chromite [4, 5]. The absolute values of the Kerr rotation angle at the extremal points are about twice as large as the corresponding values for a massive polycrystalline cobalt ferrite-chromite.

Thus, we have demonstrated that the method of solid-phase reactions in metal/oxide layer structures allows thin films of cobalt ferrite-chromite with high values of magnetic and magneto-optic characteristics to be obtained on silica substrates at relatively low temperatures of synthesis. The entire set of magnetic and magneto-optic characteristics meets requirements to magneto-optic media for MPCs and magneto-optic memory.

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