MAGNETISM AND FERROELECTRICITY

Magneto-optical Study of Granular Silicon Oxide Films with Embedded CoNbTa Ferromagnetic Particles

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Abstract—Granular silicon oxide films with embedded CoNbTa ferromagnetic particles and different relative contents of the metal and dielectric phases were studied using both steady-state and dynamic magneto-optical techniques with subpicosecond time resolution. Measurements were conducted in the spectral interval from 1.45 to 1.70 eV. The concentration dependences of the linear and photoinduced Kerr effects were found to behave similarly. Both relations are nonmonotonic with a maximum lying near the percolation threshold. © 2003 MAIK “Nauka/Interperiodica”.

1. INTRODUCTION

Granular Fe-, Co-, and Ni-based nanocomposites containing ions of other transition metals have been recently attracting considerable interest. This interest can be attributed primarily to the giant magnetoresistance (GMR) effect observed in these materials [1]. In addition, nanocomposites exhibit a number of other unusual magnetic, electrical, optical, and magneto-optical properties, among which one can place the possibility of varying the electrical resistivity within a broad range, enhancement of optical nonlinearities [2], a correlation between the magnetic transport and nonlinear optical properties [3], the high magnetorefractive effect [4], and high electromagnetic-radiation absorption in the rf and microwave regions [5]. Although some physical mechanisms responsible for these phenomena still remain unclear, granular nanocomposites can be confidently classed among materials promising for use in developing reading heads and magneto-optical storage components [6, 7]. Thus, investigation of the magnetic, optical, and magneto-optical properties of granular structures is interesting from the standpoint of both fundamental and applied research. It should be pointed out that present-day requirements for the recording of information and readout speed impose stringent constraints not only on the magnitude of magnetoresistance or of nonlinear optical susceptibility but also on the response time of a material to magnetic or light excitation.

In this communication, we report on a study of the magnetic, optical, and magneto-optical properties of granular structures of amorphous silicon oxide containing cobalt-based ferromagnetic nanoclusters with vari-

ous metallic phase contents. This investigation made use of both stationary and dynamic magneto-optical techniques, the latter providing the possibility of studying the magnetic and optical properties of a medium with subpicosecond time resolution.

2. SAMPLE GROWTH AND STRUCTURE

Granular films consisting of a silicon oxide dielectric matrix with ferromagnetic inclusions of a cobalt-, niobium-, and tantalum-based amorphous alloy with different contents of the metal phase represent typical percolation systems. Such films were prepared using ion beam sputtering on fixed glass ceramic substrates [8]. The metal grains were 2–5 nm in size, depending on the actual content of the metal phase. The metal nanoparticles made up a granular, electrically unconnected structure below the percolation threshold and a conducting cluster structure above this threshold. We studied three composites based on silicon oxide with embedded CoNbTa particles and an amorphous metal-alloy film which did not contain the oxide. The targets employed to prepare films of the metal alloy and of amorphous granular nanocomposites were of two types, namely, monolithic and composite. The monolithic targets of $\text{Co}_{86}\text{Nb}_{12}\text{Ta}_2$ composition were obtained from metals in the corresponding proportion through rf alloying in vacuum. To prepare the alloys, we employed 99.98%-pure cobalt and technical-grade tantalum and niobium, using the component weight content relative to the alloy composition required. After mechanical stirring, the melt of the corresponding composition was poured into a special ceramic mould. The

Film parameters

Sample	Number of SiO ₂ plates in target	Composition	Silicon oxide content	Thickness, μm
A	0	Co _{86.1} Nb _{12.2} Ta _{1.7}	—	5.0
B	9	Co _{53.4} Nb _{8.3} Ta _{1.1}	Si _{20.2} O ₁₇	4.6
C	11	Co _{47.3} Nb _{7.0} Ta _{1.4}	Si _{23.6} O _{20.7}	4.7
D	15	Co _{27.7} Nb _{3.0} Ta _{0.7}	Si _{26.7} O _{41.9}	4.6

melting and casting operations were performed in vacuum. Two targets measuring $270 \times 70 \times 14$ mm were melted from one charge of the alloy. The targets were ground on both sides and soldered to the water-cooled base; one of them was mounted in the vacuum chamber for subsequent sputtering. The composite target of composition Co–Nb–Ta + SiO₂ was a Co₈₆Nb₁₂Ta₂ alloy target with quartz single-crystal plates, ~2 mm thick and ~9 mm wide, fixed to the surface of the alloy target perpendicular to its longitudinal axis. By properly varying the number of quartz plates (9, 11, and 15 plates were used in our case) and the distance between them, one could change the relative volumes of the deposited magnetic and dielectric phases and, thus, control the composition of the composite. The choice of the number of quartz plates for the composite target was dictated by the need for obtaining granular composites with a unconnected and a conducting-cluster structure, as well as with a structure close to the percolation threshold [8]. The samples prepared by sputtering were films 4.6–5 μm thick. The film thickness was measured with a MII-4 Linnik interferometer. The composition of the composites thus obtained was monitored using electron microprobe x-ray spectral analysis. The parameters of the films grown are specified in the table.

3. EXPERIMENTAL SETUP

The steady-state magnetic properties of the films were studied by measuring the meridional magneto-optical Kerr effect following the technique described in [9]. We used the radiation of a femtosecond titanium-sapphire laser with photon energy $\hbar\omega = 1.45\text{--}1.70$ eV and of a cw helium–neon laser with $\hbar\omega = 1.96$ eV. The sensitivity of determining the Kerr rotation angle of the light polarization was 10 seconds of arc. The light struck the sample at 45° . The strength of the dc magnetic field applied to the sample could be varied within the interval ± 15 kOe.

Theoretical [10, 11] and, later, experimental [12] studies carried out as far back as the 1960s showed that magnetization in a medium can be produced not only by an external magnetic field but also by the electric field of a light wave. This approach, however, was earlier employed to study only diamagnetic and paramagnetic materials [13]; the investigation of magnetically ordered crystals was started only very recently [14].

Such studies make use of the completely optical technique of probing fast magnetic phenomena, in which a stronger (pump) beam transfers the medium to an excited state and a weaker beam (probe) interacts with the medium to reveal its new state. If a pulsed radiation source is available, this technique of optical pumping and probing can be employed to study the dynamic phenomena occurring in a solid with the time resolution limited by the pulse duration.

Consider a double-beam method in which a circularly polarized pump beam induces a magnetic moment in a medium as a result of the inverse Faraday effect [10, 11]. In this case, due to the Kerr magneto-optical effect, the polarization plane of the probe beam will be rotated by an angle θ_K :

$$\theta_K = \text{Im} \left(\frac{1}{n(n^2 - 1)} \int_{-\infty}^{+\infty} (p^{(+)} - p^{(-)}) E^* dt \right), \quad (1)$$

where n is the refractive index of the medium, E is the pump pulse electric field, and $p^{(+)}$ and $p^{(-)}$ are the third-order nonlinear polarizations induced by the right-handed circularly polarized pump pulse and the right- and left-handed circularly polarized components of the probe pulse, respectively [15]. We will call this phenomenon the Kerr dynamic magneto-optical effect in what follows.

Dynamic measurements were carried out using the technique described in [15]. When using a femtosecond Ti-doped sapphire laser ($\hbar\omega = 1.45\text{--}1.60$ eV) operating with a pulse duration of 100 fs and a repetition frequency of 82 MHz, the pump and probe beams were focused on a sample at a 10 : 1 intensity ratio to a spot 100 μm in diameter for the pump beam and a slightly smaller spot for the probe beam. The spatial energy exposure per pump pulse was 10 $\mu\text{J cm}^{-2}$ in most measurements. The angles of incidence of the pump and probe beams were 20° and 30° , respectively. The Kerr rotation was measured as a function of the delay time between the pulses and of the photon energy. To exclude possible spurious signals, photoinduced rotation was measured as a function of the pump polarization ellipticity. In complete agreement with the theory of the dynamic magneto-optical Kerr effect, the

Fig. 1. Field dependences of the steady-state meridional Kerr effect in films *C* and *D*.

response was maximal under circularly polarized pumping.

4. EXPERIMENTAL RESULTS AND DISCUSSION

Figure 1 displays field dependences of the Kerr effect for films *C* and *D*. The field dependences exhibit a narrow hysteresis and reach practically complete saturation in fields ± 15 kOe. The weak saturation fields imply that the CoNbTa particles embedded in the SiO_2 matrix are ferromagnetic. No noticeable dispersion was observed in the spectral responses within the photon energy range covered (Fig. 2). At the same time, one clearly sees a nonmonotonic dependence of the magnitude of the effect on the Co content, which passes through a maximum at a cobalt concentration of 45 at. %. Note that in granular films, the magnitude of the magnetooptical Kerr effect is two to three times larger than that in a metal alloy.

Investigation of the behavior of the Kerr effect with time showed the signal to be observed only in the region where the pump and probe pulses overlap. Note that in granular structures, local-field enhancement effects occurring during the pulse overlap may contribute considerably to the magnitude of nonlinear optical phenomena, in particular, of the dynamic magnetooptical Kerr effect. After termination of the pumping, the local-field effects decay rapidly as the optical coherence is lost. This may account for the dynamic Kerr effect being observed only during the pump and probe pulse overlap. Another factor that may explain this behavior of the Kerr effect is the short nonequilibrium spin polarization lifetime, which is associated with the short lifetimes of electrons in the excited state [16] and of spin excitations [5].

The spectral response of the dynamic magnetooptical Kerr effect is shown graphically in Fig. 3. In contrast to the static effect, the dynamic one reveals strong dispersion in all granular structures. The spectral response exhibits the presence of electron transitions

Fig. 2. Spectral response of the steady-state meridional Kerr effect in films (1) *A*, (2) *B*, (3) *C*, and (4) *D* measured in a field of 3 kOe. The lines plot linear fits to experimental data.

Fig. 3. Spectral response of the dynamic Kerr effect in films (1) *A*, (2) *B*, (3) *C*, and (4) *D*. The lines plot linear fits to experimental data. The inset shows (1) the steady-state and (2) dynamic Kerr effects as functions of cobalt content.

near 1.45 eV or at a lower energy beyond the operating range of the Ti-doped sapphire laser. It should be pointed out that the dynamic Kerr effect in a monolithic CoNbTa film is practically independent of photon energy. Thus, the electron transition near 1.45 eV (or at a lower energy) is obviously induced by processes occurring in CoNbTa nanoparticles and manifestations of this transition can be revealed only by using a method based on measuring the dynamic magnetoopti-

cal Kerr effect. In our opinion, the resonant behavior of the nonlinear magnetooptical signal near 1.45 eV is due to local-field enhancement effects in the vicinity of plasmon transitions in CoNbTa nanoparticles.

The inset to Fig. 3 plots the dynamic and steady-state magnetooptical Kerr effects vs. cobalt content in granular films observed at a photon energy $\hbar\omega = 1.45$ eV. The identical behavior of the concentration dependences of these phenomena appears remarkable. This implies a correlation between the steady-state and dynamic Kerr effects. Note that a correlation between the magnetic transport and nonlinear optical properties in granular nanocomposites was earlier observed experimentally in [3]. Revealing the mechanisms responsible for the interrelation of these phenomena appears to be an interesting task for further studies.

5. CONCLUSIONS

Thus, we have obtained and studied granular nanocomposites of a CoNbTa ferromagnetic alloy embedded in a silicon oxide matrix with different cobalt contents by using the steady-state and dynamic magnetooptical methods. The spectral response and concentration dependences of the steady-state and dynamic magnetooptical Kerr effects were investigated and compared. The spectral response of the linear Kerr effect exhibits a weak and monotonic dependence on photon energy. By contrast, the spectral studies of the dynamic Kerr effect provide evidence of electron transitions occurring in granular films at photon energies below 1.45 eV. The difference between the steady-state and dynamic spectral responses can be accounted for by local-field enhancement effects near the plasmon resonance in metal grains. The concentration dependences of the steady-state and dynamic Kerr effects reveal a nonmonotonic behavior and exhibit a maximum near the percolation threshold, where the cobalt concentration is about 45 at. %.

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