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Journal of Magnetism and Magnetic Materials 170 (1997) L243–L247

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Letter to the Editor

Curie temperatures of $\text{Y}_3\text{Fe}_5\text{O}_{12}/\text{Gd}_3\text{Fe}_5\text{O}_{12}$ superlattices

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Received 3 December 1996; received in revised form 29 January 1997

Abstract

The magneto-optical Kerr effects (MOKE) of epitaxial $\text{Y}_3\text{Fe}_5\text{O}_{12}/\text{Gd}_3\text{Ga}_5\text{O}_{12}$ (YIG/GGG) garnet superlattices grown on (1 1 1)GGG previously by pulsed laser deposition (PLD) were measured. A series of superlattices were investigated with the thickness of the ferrimagnetic YIG layer varied from six unit cells to only one unit cell while keeping the Curie paramagnetic GGG fixed at one unit cell. It was demonstrated that the ellipsometric technique employing photoelastic modulators (PEM) is sensitive enough to measure the MOKE signals of these ultrathin oxide samples. The Curie temperatures, determined by MOKE, are fit with a power law, yielding a shift exponent $\lambda = 3.1 \pm 1.2$.

PACS: 75.20.-i; 75.50.Gg

Keywords: Curie temperature; Yttrium iron garnet; Gadolinium gallium garnet; Magneto-optical; Kerr effect; Shift exponent

1. Introduction

The study of ultrathin metallic magnetic films is an active field where many efforts are being devoted to studying dimensionality effects, anisotropy, interfacial properties, giant magnetoresistance, and magneto-optical properties, etc. [1]. When magnetic layers become very thin, their properties depart from those of bulk materials due to new low-dimensional magnetic interactions. Although thin

magnetic-oxides are less studied, many interesting magnetic interactions are expected to occur in ultrathin magnetic oxides films. Cubic iron garnets of the formula $\text{A}_3\text{Fe}_5\text{O}_{12}$ are a large class of ferrimagnetic oxides which have applications in magneto-optical devices and magnetic bubble memories [2]. It would be interesting to fabricate ultrathin films of such oxides and to measure their properties. In this letter, we report the study of the magnetic transition temperatures of high-quality ultrathin superlattices of magnetic $\text{Y}_3\text{Fe}_5\text{O}_{12}$ (yttrium iron garnet, YIG) and paramagnetic $\text{Gd}_3\text{Ga}_5\text{O}_{12}$ (gadolinium gallium garnet, GGG) on the (1 1 1) surface of GGG substrates. To our

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knowledge, this work measured, for the first time, the thickness dependence of the Curie temperatures of ferrimagnetic oxide films down to only one unit cell ($\sim 21 \text{ \AA} = \sqrt{3}a$).

Although it is quite successful in fabricating elemental metallic films with novel structures using molecular beam epitaxy (MBE), it is relatively difficult for MBE to grow complex insulating oxides. Pulsed laser deposition (PLD) is a thin-film growth method gaining popularity after its successful application to the high-temperature superconductors (HTSC) [3]. One of the major advantages of PLD is the characteristic of congruent evaporation. Hence, complex multi-component compounds such as the HTSC can be grown rather easily by PLD [4]. Since the magnetic materials we are interested in here are also complex oxides, PLD is ideal for growing thin films of such materials.

SQUIDS are the most sensitive magnetometers available, yet the very large paramagnetic background from the GGG substrates and the high transition temperatures make it rather awkward for a SQUID to measure the Curie temperatures of our samples. Magneto-optical Kerr effect (MOKE) has recently become an important new technique to study a wide range of magnetic properties, such as magnetic anisotropy, critical phenomena, etc., of thin magnetic layers [8]. The high optical reflectivity of metallic magnetic films makes MOKE suitable for probing monolayer films. Despite the lack of conduction electrons to play the role in microscopic magneto-optics, it is equally well to use MOKE to probe insulating magnetic films, which only partially reflect the incident light. In this letter, we demonstrate that MOKE is also a sensible means to study thin insulating magnetic films down to only one unit cell.

2. Experimental

The details regarding the synthesis and characterization of the superlattices have been described elsewhere [5], and only the most relevant are given here. The superlattices were deposited on (1 1 1)GGG substrates, which were held at 650°C , by alternatively ablating a YIG and a GGG targets

with an ArF excimer laser. An oxygen flow was introduced to the growth chamber to maintain the oxygen pressure at 200 mTorr during the deposition. A series of superlattices were made with the thickness of the YIG layer varied from one unit cell to six unit cells while keeping the thickness of the GGG layer fixed at one unit cell, i.e., $[\text{YIG}(n)/\text{GGG}(1)]_m$, where $n = 1, 2, 3, 4$, and 6, and this structure was repeated m times. The surface structure of the films was examined by reflection high-energy electron diffraction. The sharp diffraction spots and highly symmetric patterns indicate that the films are epitaxial. The layered structure of the superlattices was examined by grazing angle X-ray reflectivity, which is a powerful technique for the characterization of thickness and interface roughness of multilayer films [6]. Fig. 1 shows the X-ray reflectivity of a representative sample $[\text{YIG}(1)/\text{GGG}(1)]_{12}$ and the corresponding simulated curve calculated using the software REFS [7]. Despite that there are 12 YIG layers, 12 GGG layers and 24 YIG/GGG interfaces in the sample, only three fitting variables, namely, a single thickness for all of the YIG layers, a thickness for all the GGG layers and a rms roughness for all the interfaces, are sufficient for the simulation. The results showed that the thickness of the YIG and GGG

Fig. 1. Experimental and simulated X-ray reflectivities versus grazing angle of $[\text{YIG}(1)/\text{GGG}(1)]_{12}$. The simulated curve is shifted down by one order of magnitude for comparison.

layers are both accurate within 5% of the desired thickness, and the rms roughness, $\sim 6 \text{ \AA}$, of the interfaces is no larger than that of the substrate surfaces, determined also by reflectivity, indicating that the sample is of high quality. The agreement between the experimental and simulated curves is also very good for the rest of the samples. All of the superlattices are magnetic, as measured with a SQUID, even when the YIG layer is as thin as one unit cell. A separate study showed that the magnetic properties of the sample $[\text{YIG}(1)/\text{GGG}(1)]_{12}$ do not differ from those of the sample $[\text{YIG}(1)/\text{GGG}(3)]_{12}$. Therefore, the interactions between the YIG layers are negligible and the YIG layers can be considered to be independent. Ferromagnetic resonance showed that the uniaxial anisotropy constants of the samples are negative, favoring in-plane magnetization.

The temperature-dependent magnetization of these samples was measured by the longitudinal magneto-optical Kerr effect [8], where the magnetic field was applied parallel to the films and lied in the plane of incidence of a He–Ne laser, $\lambda = 633 \text{ nm}$. Since the YIG layer is very thin and the Kerr rotation is very small, typically less than 10^{-3} degree, a modulation technique using photoelastic modulators (PEM) was employed because of its high sensitivity [9, 10]. Fig. 2 shows the experimental setup, where two modulators operated at

different frequencies, $\omega_1 \approx 50 \text{ kHz}$ and $\omega_2 \approx 55 \text{ kHz}$, were used, and the signal corresponding to the frequency $\omega_1 - \omega_2$ was measured. The reason to use two PEMs (instead of one) is to reduce the interference effects generated by the PEMs themselves, which appear right at the modulation frequencies [11]. (The noise level was found to be reduced by a factor of ten with two PEMs.) Furthermore, a complete set of the Stoke ellipsometric parameters can be measured with this method of heterodynes [12]. The temperatures of the samples were measured and controlled with a fine thermocouple attached to the surface of a blank substrate placed next to the samples. At each temperature, the signal was recorded while applying a saturating magnetic field of 2000 Oe to the samples; then, the field was reversed and the signal recorded again. Thus, the difference of the two measurements at saturation is proportional to the magnetization of the samples. In order to improve the signal-to-noise ratio, the above procedure was repeated several times for each temperature, and the data were averaged.

3. Results and discussion

Fig. 3 displays the temperature-dependent MOKE curves of the superlattices. The curve for a single YIG layer of $1 \mu\text{m}$ thick, YIG ($1 \mu\text{m}$), grown by PLD under the same growth conditions, is also shown in Fig. 3. Some oscillatory behavior of these curves is clearly seen, where the oscillations are more pronounced for samples with thinner YIG layers. Although it has not been verified quantitatively, we believe that the oscillations are interference fringes from reflections off the samples. Since YIG and GGG are semi-transparent, slightly orange, a good portion of the incident light can get through the films and the GGG substrate, then reflected (partially) by the backside of the substrate to interfere with the light reflected by the film. As the sample temperature changes, the index of refraction of GGG, and, hence, the phase difference between the two reflections also change, resulting in modulations in polarization of the total reflected light. Since the transmittance is larger for films with thinner YIG layers, the modulation is also larger for such films.

Fig. 2. Schematic diagram of the experimental setup for MOKE measurements.

Fig. 3. MOKE intensity versus temperature of (a) YIG (1 μm), (b) [YIG(1)/GGG(1)]₁₂, (c) [YIG(2)/GGG(1)]₆, (d) [YIG(3)/GGG(1)]₈, (e) [YIG(4)/GGG(1)]₆, and (f) [YIG(6)/GGG(1)]₄.

The curves in Fig. 3 do not intersect the temperature axis at large angles but bend over to form small tails, so the Curie temperatures are determined by extrapolation of the main part of the curves. The Curie temperature decreases when the thickness of the YIG layer decreases, as shown in the inset of Fig. 4. As mentioned before, the quality of the films has been carefully examined, so the lowering of T_c can be ascribed to finite size effects rather than to crystalline imperfections. It has been proposed that the critical temperature of a film of spins formed by stacking n layers of infinite-plane lattices follows a power law for large n [13].

$$\frac{T_c(\infty) - T_c(n)}{T_c(\infty)} = bn^{-\lambda}, \quad (1)$$

where $T_c(\infty)$ and $T_c(n)$ are the critical temperatures of an infinitely thick lattice and the $n \times \infty \times \infty$ film, respectively. Since YIG is a crystalline compound, the natural stacking unit is a unit cell, which contains several layers of Fe^{3+} ions [2]. Yet, if we consider the unit cell as a whole, a spin of $40 \mu_B$ [2], we may still apply the theory to the case here, with n being the number of unit cells for the YIG layer. To be a meaningful 'end point' for the thin YIG films to compare with, the infinitely thick sample

Fig. 4. Plot of $\log[(T_c(\infty) - T_c(n))/T_c(\infty)]$ for superlattices [YIG(n)/GGG(1)] _{m} as a function of $\log(n)$. The straight line is a linear fit to the data points for $n = 3, 4$, and 6 . The inset shows T_c variation as a function of n .

should be under exactly the same growth conditions. Therefore, for $T_c(\infty)$, we use the Curie point obtained from the $1 \mu\text{m}$ thick YIG sample instead of the value of a bulk YIG.

Fig. 4 shows the log-log plot of Eq. (1). A linear asymptotic fit to the plot for large n ($n = 3, 4$, and 6) was calculated to yield a $\lambda = 3.1 \pm 1.2$, and $b = 0.92 \pm 0.6$. There are about 500 unit cells of YIG in $1 \mu\text{m}$, and for $\lambda = 3.1$, the fractional shift in

T_c with respect to $T_c(\infty)$ would be $\sim 10^{-6}\%$ (or $\sim 5 \times 10^{-6}^\circ\text{C}$), which is small enough to consistently justify that YIG of $1\ \mu\text{m}$ is 'infinitely thick'. It has been predicted that λ has a value between 1 and 2 for cubic ferromagnets, depending on the models and boundary conditions assumed [14, 15]. Recently, similar experiments were reported on the finite size effects in $\text{FeF}_2/\text{ZnF}_2$ and CoO/SiO_2 multilayers, where $\lambda = 1.56$ and 1.55 were found for the Néel temperatures of the insulating antiferromagnetic FeF_2 and CoO layers, respectively [16, 17]. According to the scaling theory, λ is equal to $1/\nu$, where ν is the correlation-length exponent for the infinitely thick system [13]. Since λ is larger than 2 for the case here, it might suggest that ν is smaller for ferrimagnetic YIG so that the convergence of $T_c(n)$ to $T_c(\infty)$ is faster. However, the value of λ is found to be very sensitive to $T_c(\infty)$ with 1% change in $T_c(\infty)$ corresponding to $\sim 30\%$ change in λ . Therefore, we cannot exclude the possibility that λ lies in between 1 and 2 due to the uncertainty, $\pm 2^\circ\text{C}$, in $T_c(\infty)$.

4. Conclusions

In conclusion, we have measured the high-temperature MOKE of a series of epitaxial YIG/GGG superlattices grown on (1 1 1)GGG by PLD. In these superlattices, the YIG layers, whose thickness varies from six unit cells down to only one unit cell, can be considered to be isolated. Thus, these samples are ideal for the studies of magnetic oxides films in the ultrathin regime. It was demonstrated that the ellipsometry using two photoelastic modulators is a sensible method to measure the MOKE signals of one-unit-cell-thick YIG, which shows a magnetic transition at 125°C . The Curie temperatures of the samples, determined by MOKE, are

fit with a power law, yielding a shift exponent $\lambda = 3.1 \pm 1.2$.

Acknowledgements

MYC would like to thank Prof. C.Y. Huang for the useful discussion. This work was supported by the National Science Council, ROC under contract No. NSC86-2112-M-002-031.

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