Magnetic anisotropy and domain structure in spin-valves based on MTJ

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Abstract. We have studied the crystal structure, surface morphology, magnetic anisotropy, domain structure and tunnel magnetoresistance of spin-valves with a single and double MgO barrier layers. We have demonstrated the domain structure for soft and hard magnetic layers and observed significant changes after low temperature annealing. We have carried out magnetoresistance measurements using current-in-plane (CIP) four-probe technique and discovered a substantial difference in the values of TMR ratio for single and double MTJ spin-valves. It is shown that domain structure and magnetization reversal are the same for both systems, but otherwise the behavior of tunnel magnetoresistance is different, because of second MgO barrier effects on the system conductivity. The ability to manipulate the magnetization direction in MTJ systems using temperature annealing is demonstrated. It makes these structures possible for new applications in nanoelectronics as magnetic recording media and high sensitive sensors.

1. Introduction

The multicomponent magnetic systems are new class of spintronic materials having unique combination of magnetic and electrical properties for prospective nanoelectronic applications as a non-volatile magnetoresistance memory (MRAM), spin-valve sensors and microwave spin-current generators [1-3]. The spin-valves based on magnetic tunnel junctions are very interesting objects for investigation, because of they combine all remarkable properties of both devices: high magnetic field sensitivity and high value of magnetoresistance [4,5]. The classical spin-valve is based on two ferromagnets with different coercive forces separated by a thin metal spacer [5]. The relative magnetization orientation of the two ferromagnetic layers strongly modulates the charge flow through the structure. Using this property of the spin-valve, external magnetic field acting on the magnetic moments of the ferromagnetic layers can be sensed electrically. Spin-valves with insulating interlayers, such as Al\textsubscript{2}O\textsubscript{3} [6] or MgO [7], are called magnetic tunnel junctions (MTJs) and designed to have two stable parallel and anti-parallel states, can also serve as non-volatile solid-state memory elements [8]. These applications, as read head sensors in hard magnetic disks and memory cells in MRAM are perfect examples of a fast transformation of a fundamental physics discovery [9–11] in to large scale practical devices. This makes spintronic nanostructures not only of fundamental interest,
but also of great technological concernment [3, 12]. Recently the MTJs with double tunnel barrier [13] have began to attract an attention of scientists and engineers due to be an effective means to improve the bias dependence of the tunnel magnetoresistance (TMR) ratio [14] and to reduce the critical current density of spin torque transfer [15]. Moreover, for double barrier MTJs the exciting phenomena such as quantum confinement effect, Coulomb blockade and spin-dependent resonant tunnelling have been predicted and observed [16,17].

The most effective method to improve MTJs' properties is a thermal annealing in vacuum. An annealing can generate better interfaces between magnetic layers and insulating barrier. It is important to find optimal annealing parameters to produce MTJs with clearly defined spin-valve character and high TMR ratio.

The main purpose of this paper is to study temperature dependence of magnetization reversal and an effect of thermal annealing on magnetic and magneto-transport properties of spin-valves with single and double insulating MgO barriers.

2. Experimental details

The structures Ti(30)/Au(20)/Co(20)/MgO(2)//[Co(0.3)/Pt(0.8)]_5/MgO(x)/Au(5), where x is 0 (single magnetic tunnel junction – SMTJ) or 2 nm (double magnetic tunnel junction – DMTJ), were fabricated on natural oxidized single crystal Si square substrates (1 × 1 cm^2) with an electron-beam deposition system in vacuum 10^{-7} Torr. The layers’ thickness is shown in nanometers. The top Au layer is exploited as a cap layer to prevent metal oxidation. The surface morphology and domain structure were studied with atomic force microscope (AFM) and magnetic force microscope (MFM). Transmission electron microscopy (TEM) was exploited to observe a grain size. Hysteresis curves were obtained by longitudinal magneto-optical Kerr effect and with vibrating sample magnetometer (VSM). The magnetic measurements were done at room and low temperatures using SQUID magnetometer. TMR ratio was measured with four-probe station in current-in-plane (CIP) geometry [18] at different configurations: I⊥H, I⊥n, H⊥n (transverse TMR) and I∥H, I⊥n, H⊥n (longitudinal TMR), where I is an applied current (100µA), n is a perpendicular to the surface plane, H is an applied magnetic field. Temperature annealing was performed in commercial annealing setup in vacuum 10^{-2} Torr during 15 minutes at each temperature.

3. Results and Discussion

The TEM images have shown that all as-deposited samples have polycrystalline structure with an average grain size 6 nm. An average amplitude and period of roughness are h=1,5 nm and l = 40 nm, correspondingly. The VSM measurements have indicated that our films possess an in-plane magnetic anisotropy only.

The temperature dependence of the coercive forces of hard magnetic (Co/Pt)_5 stack $H_c^{(1)}$ and soft magnetic cobalt layer $H_c^{(2)}$ for SMTJs is represented in Fig.1. For DMTJs we have obtained the same curves. If measurement temperature is dropping from 293 K to 4.2 K, the coercive force $H_c^{(2)}$ changes from 35 Oe to 40 Oe. The enhancement of $H_c^{(2)}$ is due to a contraction of thermal fluctuations of magnetic moments. $H_c^{(1)}$ changes significantly. At room temperature (RT) $H_c^{(1)}$ is 350 Oe and it increases in three times (to 1kOe) at 4.2 K. It is known, that $H_c=2K/M_s$, where $M_s$ is saturation magnetization and $K$ is an anisotropy constant. Therefore, the behavior of $H_c^{(1)}$ can be explained by the change in the values of anisotropy constant and saturation magnetization at different measurement temperatures. According to [19] the anisotropy constant increases in about 6 times in temperature interval from RT to 4.2 K, but $M_s$ in Co/Pt system grows in about 1.3 times only [20].
Figure 1. The hysteresis loops of MTJ in dependence on measurement temperature obtained with SQUID magnetometer.

We have studied an angular dependence of coercive force for both magnetic layers at RT. The dependences $H_C = f(\alpha)$ for SMTJs and DMTJs, $\alpha$ is an azimuth angle (this angle was chosen between one side of the square sample and the direction of an applied magnetic field), were measured with the longitudinal magneto-optical Kerr effect, Fig. 2. The coercive force of hard magnetic ($Co/Pt)_5$ multilayer stack $H_C^{(1)}$ is 350 Oe and for bottom soft magnetic Co layer $H_C^{(2)}$ is 35 Oe. As shown in Fig. 2, the minimum value of $H_C^{(1)}$ repeats itself every 180°. At the same time the easy axes of soft and hard magnetic layers are displaced on 90° relatively to each other. The same behavior has been observed for SMTJs. This behavior of the coercivity can be explained by the following. By the reason of high roughness of the layers ($h = 1.5$ nm for as-deposited films) we suggest, that thin MgO interlayer is structurally nonsolid (i.e. it has fluctuation of thickness). Then, at the places, where MgO thickness less than 1.0 nm, the realization of an indirect exchange coupling between soft and hard layers is possible [21]. According to Slonczewski [22] the fluctuations of interlayer’s thickness can lead to an indirect biquadratic exchange coupling between layers. Another possible mechanism of biquadratic coupling is the ferromagnetic pinhole formation through the non-magnetic interlayer [23]. Biquadratic exchange coupling leads to a perpendicular alignment of magnetic moments in adjacent ferromagnetic layers, as we have found in our as-deposited films, Fig. 2.

Figure 2. The angular dependence of the coercive force for DMTJs. The empty dots and filled squares are related to the hard and soft magnetic layers, correspondingly.
After annealing of the samples at $T_{\text{ann}}=250^\circ \text{C}$ the coercive fields $H_c^{(1)}$ and $H_c^{(2)}$ increase on 80% and 29% correspondingly. The coercive force in polycrystalline films is caused by the domain wall pinning on grain boundaries, by dispersion of magnetization of easy axes and by surface roughness. During annealing the grain size increases. This is the reason for rise of the coercive force component responsible for dispersion of magnetization of easy axes in multilayer film [24]:

$$H_{c,k} = 1.1 \frac{K_i 4^{1/3} R 4^{1/3}}{\mu_0 M_s \delta^{1/3}},$$

where $K_i$ is an induced anisotropy, $R$ is an average grain size, $\delta$ is a domain wall width, $\gamma$ is an energy density of a domain wall. After annealing at $T_{\text{ann}}=250^\circ \text{C}$ the average grain size increases in 4 times. This is the result in growth of $H_{c,k}$ in 6 times. It explains the increase of the coercive force in both magnetic layers after annealing.

As shown in Fig.3, with increase of the annealing temperature the easy axis of soft magnetic layer is rotated and aligned in parallel with easy axis of hard magnetic layer. The turn of the easy axis of soft layer occurs under influence of the magnetostatic field of the hard layer during annealing. We did mention significant influence of further annealing at higher temperatures on magnetic anisotropy in the films. As shown in [25], temperature annealing in layered magnetic systems can affect on interface roughnesses, hereupon the strength and type (ferromagnetic or antiferromagnetic) of an exchange coupling is changing. As result, biquadratic exchange coupling can decrease and even vanish after annealing. It means that magnetostatic interaction becomes to play major role in alignment of magnetic moments in adjacent soft and hard magnetic layers.

The difference in coercive forces for [Co/Pt]$_5$ and Co layers allows to observe the domain structure of soft and hard magnetic layers separately by applying different magnetic fields to the samples. Similar investigation is slightly unique, because the partition of domain structure layer-by-layer is a non-trivial task. The analysis of MFM images, received at step-by-step incremented magnetic field in range from -600 to +600 Oe, has displayed the big difference in the size and the shape of magnetic domains for soft and hard layers, Fig. 4.

At the demagnetized state the size of domains in soft layer is about 30-60 um, Fig.4(a). If to magnetize sample from 0 to the saturation field (600 Oe) and go back to -50 Oe, we have found very complicated domain structure of the hard layer, like in films with perpendicular anisotropy, Fig. 4(b). The domain size is 0.5-1 um. After annealing at $T_{\text{ann}}=150^\circ \text{C}$ the domain structure of soft magnetic layer changes appreciably, Fig. 4(c). The domains become smaller. Domain walls are extensive and unilateral. It was an issue to receive MFM image of domain structure of hard magnetic layer, because after annealing the surface roughness of the film increased substantially. This is a reason why we have done domain imaging just for samples after low temperature annealing.
Figure 4. MFM images of domain structure for as-deposited (a, b) and annealed (c) samples. (a) – at magnetic field $H = H_C^{(2)}$, (b) – at $H = -50$ Oe, (c) – at magnetic field $H = H_C^{(2)}$. The scale is the same for all images.

Using the magneto-optical Kerr effect magnetometer, we have measured hysteresis loops at RT for SMTJs and DMTJs. The behavior of magnetization reversal for SMTJs before and after thermal treatment is shown in Fig.5. The characteristic steps on the hysteresis loops prove the spin-valve nature of our samples. After annealing the coercive force of the soft magnetic layer changes slightly, but coercive field and relative magnetization of the hard layer become larger, due to increasing of grain sizes. The magnetization reversal in DMTJs has the same behavior. Due to the different coercivity of soft and hard magnetic layers, the magnetization switching occurs at variant applied magnetic fields. As seen in Fig. 5 there are two mechanisms during switching process: spin-flip in soft layer at low magnetic field (30-35 Oe) and domain wall displacement in the hard layer at higher applied magnetic fields.

Figure 5. The typical hysteresis loop of as-deposited and annealed spin-valves based on SMTJs.

To study an influence of crystal structure modification on TMR ratio we measured magneto-transport properties of our samples before and after annealing in vacuum. TMR curves, proving spin-valve behavior and spin polarized tunneling of samples, are represented in Fig. 6 and 7. We have measured TMR for as-deposited samples in longitudinal and transverse geometries at RT. TMR ratio has been defined as $(R_{AP} - R_P)/R_P$, where $R_{AP}$ and $R_P$ are the resistances for anti-parallel (AP) and parallel (P) alignment of the magnetic moments, respectively [26]. The difference between the
coercive forces of soft and hard layers allows to easily form the parallel and antiparallel magnetization states by changing an external magnetic field. This is the essential property of a spin-valve structure. The scheme of a spin-valve in Fig. 6 demonstrates magnetization switching process at change of applied magnetic field from negative to positive value. If go in backwards, we usually obtain symmetric TMR curves. If we apply negative saturation magnetic field, we get small resistance compare to the high resistance state, when the magnetization in the hard and soft magnetic layers aligned antiferromagnetically. Due to the spin-dependent tunneling, the TMR curves have a peak, indicating the maximum value of TMR ratio. In SMTJs the longitudinal and transverse TMR ratios are 20% and 17%, correspondingly. We have found bigger TMR ratio for SMTJs, than for DMTJs. In DMTJs these values are 7% and 12.5% accordingly, Fig. 7. Apparently, the second MgO barrier decreases the tunnel conductivity of the DMTJs, which is subject to less TMR ratio [27].

![Figure 6. The longitudinal (solid line) and transverse (dash line) TMR in SMTJs.](image)

![Figure 7. The longitudinal (solid line) and transverse (dash line) TMR in DMTJs.](image)

In Fig. 8 the affect of the thermal treatment on magnetoresistance is shown. In our MTJs TMR ratio is expected to decrease, because the presence of defects like interface roughness, pinholes, interdiffused interfaces, impurities or vacancies, or stacking faults, would provide additional conduction channels. There is no interdiffusion in Co/Pt stack after annealing at low temperatures [28]. Thus, the most likely reason in the decrease of TMR ratio is pinholes formation in MgO layers [21], due to current leakage through them. The TMR ration for DMTJs changes slightly, because of smaller initial tunnel conductance of electrical current, passing through two MgO barriers.
4. Conclusions
We have presented the recent results on spin-valves with single and double MgO barriers. We have studied magnetization reversal at low temperatures and magnetic and magneto-transport properties after thermal annealing. The investigation is shown, that using temperature annealing we can fabricate a spin-valve structure with predefined TMR ratio and tailored direction of magnetization in soft magnetic layer. The represented results demonstrate the possibility to design novel devices for magnetic memory and programmable logic.

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