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Effect of the target temperature during magnetron sputtering of Nickel

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ABSTRACT

By disconnecting a nickel target from a water-cooled magnetron, thereby inducing a rise in the target temperature, the time evolution of the cathode voltage and of the magnetic field above the target was measured using a small Hall-effect sensor, enabling the ferromagnetic – paramagnetic transition of the Ni target to be clearly identified. A heat flux sensor was implemented in the high vacuum chamber to measure the energy transferred to the substrate, especially the energy contribution coming from the infrared radiation emitted by the heated target. By coupling the heat flux and the Hall sensors, this study shows that the emissivity and the temperature of the target surface can be determined. A temperature up to 720 ± 30 °C and an emissivity of 0.4 ± 0.02 were obtained for a specific power density of 5 W cm^{-2} applied on the target. Nickel thin films were deposited on silicon at low and high temperatures (target respectively connected to and disconnected from the magnetron). It was found that the increase in the target temperature leads to an increase in the deposition rate (15% enhancement in the present conditions) and a change in the Ni thin film microstructure (toward a dense and compact columnar structure).

1. Introduction

Magnetron sputtering (MS) is a well-known plasma coating process commonly used in research and industry since the invention of the planar cathode arrangement credited to J.S. Chapin with a patent filed in 1974 [1]. Almost all metallic materials can be sputtered, oxides and nitrides can be synthesized, the deposits present excellent uniformity and reproducibility, the coating surface is very smooth (no droplets), and the cathodes (up to a few meters long) can be adjusted with different geometries in order to coat various shapes of substrate [2]. All these advantages make magnetron sputtering a highly flexible deposition process. However, it suffers from some drawbacks: the deposition rate is low compared to atmospheric (physical and chemical) deposition techniques, and magnetic materials are difficult to sputter (the magnetic field delivered by the magnetron is screened by the target). The quality of the thin films, even if satisfactory for many applications, in some cases requires improvements in terms of density and crystallinity. Quality can be improved by using high power impulse magnetron sputtering (HiPIMS) [3–6] or by promoting an increase in the target temperature by decreasing the efficiency of the water cooling. For example, Yang et al. [7] showed that with a hot target the formation of a better crystallized transparent and conductive aluminum-doped zinc oxide is promoted. Kawamata et al. [8] showed that the process induced the formation of stoichiometric MgF_2, whereas non stoichiometric films were obtained in conventional MS. In addition to the change in the composition and quality of thin films, the sputtering of a hot target may also induce a higher deposition rate. This result was first observed by Chau et al. in 1996 for silicon and niobium oxides [9]. Moreover, the hot target leads to a reduction of the hysteresis loop in reactive sputtering. These results have been confirmed by several groups for other oxide and nitride thin films [10–12]. Although the use of a hot target appears to have many advantages, the process is challenging to control, in particular because the target temperature is difficult to estimate without invasive tools such as thermocouples [13]. As a first approximation, the maximum temperature that could be reached can be estimated by assuming that the electrical power is dissipated in the heating process of the target as in Ref. [14]. With this model where 95% of the electrical power is assumed to be lost in radiative transfer, the maximum temperature was estimated with the Stefan–Boltzmann law. By applying this

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model to a circular target (1 mm thick, 2 inches in diameter) with an emissivity of 0.4, the temperature reached 665 °C at 50 W (specific power density of 2.5 W cm^{-2}). A more accurate approach to estimate the target temperature is to simulate the whole process of thermal transfer based on the energy balance in the "magnetron/target" system. Bleykher et al. [15] give the temperature of a Cr target thermally disconnected from the magnetron (graphite crucible + insulating gap) in stationary MS. The temperature ranged between 800 and 1200 °C for a specific power density between 5 and 20 W cm^{-2}, considering the power evacuated by heat radiation from the target surface (+crucible), by the ions reflected from the target and by the sputtered atoms (and possibly evaporated ones). To obtain a precise measurement, Tesar et al. [16] used an infrared (IR) camera and the phase transition of the Ti target (from β-to α-phase at 882° C) powered in HiPIMS regime to estimate the emissivity (0.28) and to evaluate the surface temperature. A temperature up to 1750 °C was measured on the 4-inch Ti target with a peak power of 35 kW (peak current of 35 A, 1 kV, 445 W cm^{-2}) and a 20% duty cycle (mean specific power density of 89 W cm^{-2}). All these studies show that the target surface can be significantly heated and that the estimation of the temperature attained is not an easy task.

In this study, we used an energy flux diagnostic based on a heat flux microsensor (HFM) [17] to study the energy delivered to the substrate during magnetron sputtering. This tool can detect IR radiation emitted from the target [18]. A compact Hall-effect sensor was used to measure the magnetic field above a nickel target for which the magnetic transition is observed at the Curie temperature T_c (T_c = 358 °C). Thanks to the T_c identification coupled with the IR radiation measurement, the emissivity of the Ni target can be evaluated. The time evolution of the target surface temperature can therefore be estimated by assuming that emissivity ε remains constant. Finally, the effect of the target temperature on the deposition rate and microstructure of Ni thin films is evidenced.

2. Experimental section

All experiments were performed in a 6-way cross-shaped high vacuum (HV) deposition chamber pumped by a 500 l s^{-1} turbomolecular pump (Pfeiffer 400M) and a primary pump (Adixen ACP15) down to 5 10^{-4} Pa. A directly cooled 2-inch magnetron (ONYX-02, Angstrom science) is horizontally fixed on a 200 ISO-LF flange. A custom-made magnet assembly was specifically designed from 21 commercial cylindrical NdFeB magnets (Ø 6 mm, 8 mm height) in order to have a high magnetic field. Fig. 1a gives the components of the magnetic field (tangential $B^{//}$and normal $B^{\perp}$) measured 11 mm above the magnetron backing plate. The pressure is regulated to 1 Pa after the introduction of Argon gas with a flow rate of 10 sccm. The plasma is ignited by a Pinnacle Plus power supply (Advanced energy, 5 kW) in DC mode regulated at 25, 50, 100, 150 and 200 W. For each power value, the time evolution of the voltage is monitored on a data acquisition system (DAS 600, Sefram) through a high voltage probe (P6015A Tektronix). A 1 mm thick Nickel target is mechanically clamped on the magnetron. In order to promote the target temperature increase, a 1 mm thick Macor® ceramic disk is inserted between the magnetron and the target which allows thermal disconnection while preserving the electric contact via the annular clamping fixed on the magnetron body by 4 metallic screws. This magnetron configuration, denoted HiTeMS in this study for High Temperature Magnetron Sputtering, can lead the target to glow red as shown Fig. 1b. The ceramic disk was removed in some experiments in order to compare HiTeMS and conventional MS configurations.

The magnetic field is qualified using a small, linear Hall-effect device (AH49F 4 × 4.2 × 1.6 mm, diodes) placed 11 ± 2 mm above the magnetron backing plate in front of the racetrack (about 15 mm from the center) in order to be sensitive to the $B^{//}$component. The distance between the target and the sensor is respectively 10 ± 2 mm and 9 ± 2 mm in the HiTeMS and in MS configurations considering the thickness of the target and of the ceramic disk. The sensor is placed inside a 50 mm long Macor® tube, one end of which is covered by Kapton® tape in order to protect the sensor from Ni deposition. The voltage delivered by the sensor varies in proportion to the strength of the magnetic field with a sensitivity of 2 mV $Gauss^{-1}$. As the sensor is temperature-sensitive (range of temperature: −40/+105 °C), the Macor tube is placed on a manual linear manipulator and the sensor is moved back between each measurement spaced by 10 s (0.1 Hz). The target temperature is estimated from the emitted IR radiation measured with an energy diagnostic composed of a commercial heat flux microsensor (Vattell-HFM-7) according to a procedure previously described in Ref. [18]. The HFM is composed of a thermopile sensor (approx. 6 mm in diameter) which provides a voltage variation directly proportional to the incoming energy flux density (Φ). It is calibrated according to a NIST protocol based on IR radiations emitted from a black body. The HFM voltage was registered every 0.5 s using a nanovoltmeter (Keithley 2182). A 6 mm diameter copper substrate was glued on the HFM using thermally conductive paste. The HFM is water-cooled and its temperature is fixed to 5 °C. This is required to avoid reaching an equilibrium which would lead to the balance of the in and out energy fluxes. For detailed conditions of HFM use, the reader is referred to [17]. Just before the experiments, the HFM is placed at a distance of 60 mm from the target center and displays an energy flux density of 13 mW cm^{-2} (Φ_{offset}). In order to evaluate the temperature of the target surface through the measurement of the energy flux density, the emissivity of the target surface must be known. The identification of a phase transition during the measurement can be advantageous to evaluate the emissivity as reported in the study [16] on the sputtering of a titanium target (solid state transition from β-Ti to α-Ti at 882 °C). Here, the magnetic transition at the Curie temperature (358 °C) was evidenced by the Hall-effect sensor measurements. From the measurement of the IR contribution at this step, the temperature being known, it is possible from the Stefan-Boltzmann law to evaluate a mean emissivity of the target surface. Nickel thin films were deposited on (100) Si substrate at various powers in conventional MS and in HiTeMS configurations for a substrate/target distance of 60 mm, an Argon pressure of 1 Pa and two deposition times of 5 and 20 min. Rutherford Backscattering Spectroscopy (RBS) was used to measure the precise amount of Ni atoms deposited on the Si substrates. For RBS measurements, the energy of the probing ^{4}He particles and the scattering angle were fixed to 2 MeV and 165°, respectively. Finally, the microstructures obtained in MS and in HiTeMS configurations were compared by observing the Ni thin film cross section by scanning electron microscopy (Zeiss Supra 40).

3. Results and discussion

Fig. 2a displays the time evolution of the magnetic field measured by the Hall-effect sensor for 50, 100 and 200 W in the HiTeMS configuration. In the absence of plasma, the $B^{//}$component above the target reaches 165 G in both configurations as shown on the graph. Just after plasma ignition, the magnetic field increases from 165 Gauss to about 240 Gauss in the HiTeMS configuration. This maximum value is obtained after 25, 50 and 100 s for the respective powers: 200, 100 and 50 W. At the maximum value, the nickel target no longer screens the magnetic field delivered by the magnetron. This value, 240 Gauss, is slightly lower than the maximum $B^{//}$value shown on Fig. 1a. One reason may be that the sensor is not placed exactly at the position where $B^{//}$is maximum in