UHV plasma jet system for deposition of magnetic nitride nanocomposite films with GHz applications

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Abstract. A method of preparation of extremely pure magnetic thin films, especially magnetic nitride nanocomposites for GHz applications was searched. The plasma-jet method was chosen for its advantages at magnetic materials deposition. Sources of impurities deteriorating the quality of the films were analysed. Based on the assumption that the achievable purity is limited mainly by the conditions at the deposition, an experimental UHV apparatus with the plasma-jet was designed. A number of magnetic thin films from various materials including nitride nanocomposite films was prepared already in this apparatus at UHV conditions. Their magnetic properties are far better than those of the films prepared in a high vacuum apparatus.

1. Introduction

Magnetic thin films formed from pure metals, alloys or compounds with defined structures are prospective materials for many applications. Thus, also effective methods of their preparation enabling to reach optimum parameters have to be found. The layers of complex structure and composition have to be sputtered. Magnetic field is applied in the space of deposition at magnetron sputtering but the growing magnetic layer draws the field and it affects the operation of the magnetron and vice versa the non-uniform magnetic field has undesirable effects on the growth of the layer.

A plasma-jet deposition (hollow cathode discharge) is an advantageous alternative to the magnetron sputtering [1]. The jet is in a vacuum chamber directed against the substrate. Its nozzle is manufactured from the material to be deposited. Inert working gas (argon) flows through the jet into the chamber and it is pumped out with high effective pumping speed. The discharge (either DC or RF) glowing inside the jet sputters its material and the gas stream carries the sputtered particles on the substrate. In principle no magnetic field is necessary and the speed of deposition is comparable with that of magnetron sputtering. Complex alloys can be deposited or even produced only during the deposition from single materials inserted into the jet nozzle.

Both the magnetron sputtering and the plasma-jet deposition are performed in vacuum. Albeit the average pressure in the chamber during the deposition is in the range from some tenth Pa to some hundreds Pa (the pressure distribution is always highly non-uniform), the partial pressures of some gases have to be very low, because these gas species enter the discharge and they react with the...
deposited materials. The formed compounds are embedded into the layers and they strongly affect the magnetic properties. The gases causing formation of oxides are one example.

2. The impurity sources at magnetic film sputtering

Only pure inert working gas has to be present in the discharge space during the film deposition. Actually, the following sources of undesirable gases can take effect:

a) impurities in the source working gas;

b) impurities admixed into the working gas in the admission lines, valves and so on;

c) the gas dissolved in the sputtered material or fixed in chemical compounds contained in this material;

d) the gas leaking into the apparatus;

e) the gas released spontaneously from inner surfaces (gassing);

f) the gas released during the deposition due to the interaction of the discharge with the deposition device and with the walls of the chamber (surface particle bombardment, heating effect, radiation);

g) the gas from the pump back-stream.

Only the a) and c) items depend only on the quality of the used working gas and the material to be sputtered. All the others are strongly influenced by the condition of the vacuum apparatus. In order to suppress these gas sources the apparatus and the admission lines have to be entirely vacuum tight. The gas admission lines have to be all-metal enabling to pump out at elevated temperature and to flush with dry working gas. The parts of the apparatus subjected to the attack of high temperature, charged particle bombardment and radiation during the jet operation have to be vacuum annealed before the apparatus assembly. The type and size of the pump has to be properly chosen with regard to the plasma jet function also. In order to suppress the spontaneous gas release from the inner surfaces, the film of adsorbed gas restored at each opening the chamber has to be removed before the start of the film deposition.

3. The difference between HV and UHV apparatuses for magnetic film sputtering

One of the gases the content of which in the layers must be carefully controlled is oxygen. Oxygen gas can be hardly present in a vacuum apparatus free of leak but the source of oxygen in the discharge is water desorbing from the layer adsorbed on all inner surfaces. A simple estimation can illustrate its role: A pressure of the order of $10^{-5}$ Pa can be achieved in a leak-free HV chamber within several tens minute but the subsequent pressure decrease is only very slow. The vast majority of the residual gas is water. If, for example, a pure iron layer has to be deposited with the deposition rate $10$ nm/min then $1.4 \times 10^{19}$ atoms of iron impact on $1$ m$^2$ per $1$ s together with several times $10^{17}$ molecules of water each carrying one atom of oxygen. Thus, the content of oxygen in the layer cannot be less than some atomic per cent though the sputtered material was perfectly pure.

The most reliable way to ease off the adsorbed water is to pump the apparatus for several hours at elevated temperature (at least 250 °C in the case of stainless steel chamber). The apparatus has to be designed as an UHV apparatus for this purpose. All the materials and the design concepts notwithstanding the baking out have to be given out. This makes the design of the apparatus providing all options of the plasma-jet technique (water cooled jet, alternatively cooled and heated sample holder, transfer of the sample holder between various positions, mutual positioning of the jet and the substrate using both direct and convergent jets without vacuum break) difficult.

4. Experimental UHV apparatus with plasma-jet

The plasma-jet technique enables to produce complex layers and to influence their structures and properties many ways. A versatile UHV apparatus with plasma-jet was designed with the two aims:

- to prove the influence of the UHV conditions on the quality of the produced films;
- to provide option to set up various parameters of the process in wide range in order to follow the influence on resulting film parameters.
The apparatus is equipped in stages with UHV compatible equipment to enable without vacuum break:

- To adjust the distance between a direct jet and the substrate. The substrate temperature is stabilised (substrate either cooled or heated).
- To transfer the substrate between the positions against two direct jets at multilayer deposition.
- To use one pair of convergent with variable the distances between each convergent jet and the point of convergence and the distance between the point of convergence and substrate mutually independently.
- To clean the substrate by means of ion bombardment and to evaporate the sub-layers on the substrate.

Because the properties of the deposited films are influenced also by the average gas pressure in the chamber at a given gas throughput [2] also it can be adjusted in wide range. That is why a pump with high pumping speed (1500 ℓ/s) was used. The effective pumping speed is adjusted by means of a gate valve in the ratio \( S_{\text{EFFMIN}} : S_{\text{EFFMAX}} = 1 : 1000 \). Dry pumps were preferred. That is why a cryogenic pump was chosen as the main pump.

5. Results and discussion

The UHV apparatus, the UHV compatible water cooled plasma jet and the UHV compatible adjustable substrate holder alternatively cooled or heated were developed successfully. The expected vacuum parameters were achieved. The ultimate pressure of the chamber before start sputtering is typically 2-3 \( \times 10^{-7} \) Pa. The partial pressure of water is usually approximately 1×10^{-8} Pa.

The UHV plasma-jet-deposition apparatus is successfully used for depositions of special magnetic films based on 3d-metals. These materials consist of elements with very high affinity to oxygen. The first successful experiments with deposition of pure iron Fe and aluminium Al coatings, preparation of pure dielectric AlN film and stoichiometric soft magnetic mumetal alloy Ni_{73}Fe_{15}Cu_{7}Mo_{4}Mn_{1} films showed that addition of oxygen in these films is not higher than 0.02 atomic % [3].

Magnetic nitride nanocomposite FeCo-AlN films with GHz applications [4, 5] were finally prepared [6]. Plasma deposition process was performed by reactive sputtering of combined Fe_{50}Co_{50}+Al nozzle in Ar + N_{2} working gas mixture flow (total pressure 0.15 Pa, partial N_{2} pressure 0.35%) on water-cooled (at 15°C) Si, SiO_{2}(200 nm)/Si and glass substrates. Magnetic anisotropy of films was induced by applied external magnetic field during the deposition. The investigated films have thickness about 600 nm and the approximate composition Fe_{40}Co_{40}Al_{10}N_{10}. X-ray diffraction and chemical analysis revealed that the films consist of nanocrystalline FeCo grains with the size of about 10 nm and dispersed amorphous AlN clusters. Magnetic domain structure refinement in hard-axis fields and on the edge is typical for uniaxial thin films. Samples were heat annealed at 300 °C for 1 hour in a magnetic field of 50 mT. Resulting materials are magnetically soft with coercivity about 0.4 mT (Fig. 1). The high frequency permeability dependence was investigated in the range of 0.1–5.0 GHz. The characteristic frequency at which the natural ferromagnetic resonance occurred is about 2.4 GHz (Fig. 2). The resistivity is increased by the nitridation from original value \( \rho \approx 40-60 \mu \Omega \text{cm} \) up to 160-200 \( \mu \Omega \text{cm} \), thus, the eddy currents are distinctively suppressed in the films at their use in the microinductors for GHz mobile communication devices.

These magnetic parameters of the films are much better than those achieved before with a high vacuum apparatus.

6. Conclusions

The assumption that the vacuum conditions in a HV apparatus at the magnetic film deposition are the main reason limiting achievable purity, low content of oxygen and thus also magnetic parameters of the films proved right.

A versatile UHV apparatus with plasma-jet was designed. Main devices necessary for plasma-jet technique deposition were developed as UHV compatible.
The UHV plasma-jet-deposition apparatus is successfully used for depositions of special magnetic films based on 3d-metals and magnetic nitride nanocomposite FeCo-AlN films with GHz applications. The magnetic parameters of these films prepared in UHV conditions surpassed considerably the parameters of films prepared in HV conditions.

![Hysteresis loops of FeCo-AlN nanocomposite film prepared by the UHV plasma jet (coercivity 0.4 mT in Easy Axis).](image1)

**Figure 1.** Hysteresis loops of FeCo-AlN nanocomposite film prepared by the UHV plasma jet (coercivity 0.4 mT in Easy Axis).

![HF ferromagnetic resonance (~2.4 GHz) measurement of the same FeCo-AlN sample.](image2)

**Figure 2.** HF ferromagnetic resonance (~2.4 GHz) measurement of the same FeCo-AlN sample.

7. References

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