Thermal stability of the ferromagnetic in-plane uniaxial anisotropy of Fe-Co-Hf-N/Ti-N multilayer films for high-frequency sensor applications

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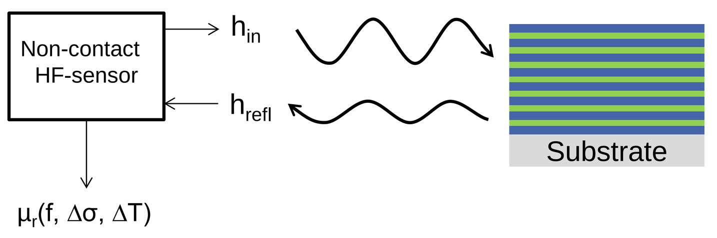
Motivation

Idea: contactless inspection of wear state of surfaces and coatings

Ferromagnetic material — Fe-Co-Hf-N

Protective material — Ti-N

multilayer design: protective coating with an integrated sensor function

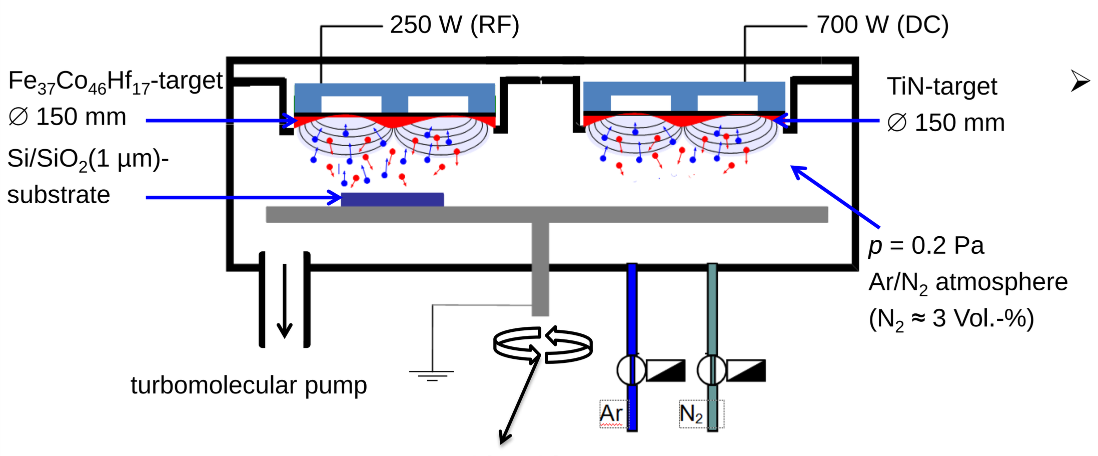


Quantification of mechanical stress changes (Δσ) and temperature changes (ΔT) in the sensor material by a shift of the resonance frequency f_r

Sample preparation

➢ Two-step process: Deposition and subsequent heat treatment

1. Reactive DC and RF magnetron sputter deposition



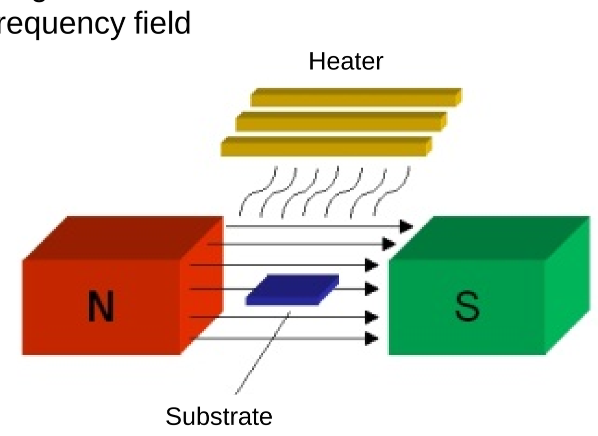
Rotatable table:

- $Fe_{32}Co_{44}Hf_{12}N_{12}$ and $Ti_{50}N_{50}$ individual layers with number of bilayers n = 7
- Individual layer thickness $Fe_{32}Co_{44}Hf_{12}N_{12}$: $d_{FeCoHfN}$ = 53 nm
- Individual layer thickness $Ti_{50}N_{50}$: d_{TiN} = 67 nm

2. Heat treatment

- Annealing for 1 h at T_a = 400 °C or T_a = 600 °C in a static magnetic field (50 mT) in vacuum after deposition

➢ Generation of an in-plane uniaxial anisotropy H_u to ensure a homogenous precession of magnetic moments in an external high-frequency field



Experimental

Temperature-dependent ferromagnetic properties

- A shift in the resonance frequency f_r with increasing temperature is expected due to a decrease of J_s
- Temperature stability of the resonance peak depends on a possible degradation of thermally induced uniaxial anisotropy field H_u at high temperatures

Study:

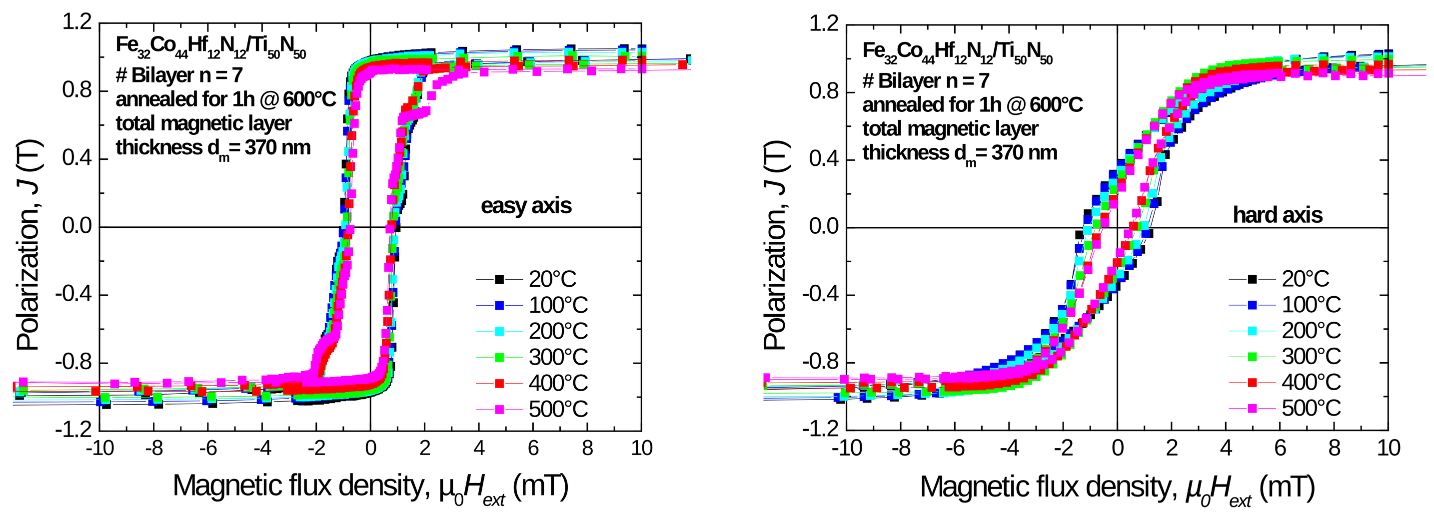
- **Investigation of the thermal stability of H_u thermally induced at T_a = 400 °C and T_a = 600 °C**

➢ **Temperature-dependent VSM measurements in easy and hard axis of polarization from room temperature (RT) up to 500 °C**

Results

Multilayer films annealed for one hour at T_a = 600 °C in vacuum

Temperature-dependent hysteresis loop measurements in easy and hard axis of polarization from RT up to 500 °C in air



- Decrease of coercive field H_c in the hard axis of polarization
- Saturation polarization J_s decreases with increasing temperature
- Clear distinction between easy and hard axis up to 500 °C
- Absolute value of μ_0H_u decreases slightly from 5 mT at RT to 3.4 mT at 500 °C
- **Uniaxial anisotropy field μ_0H_u remains stable in its direction up to 500 °C within one hour**

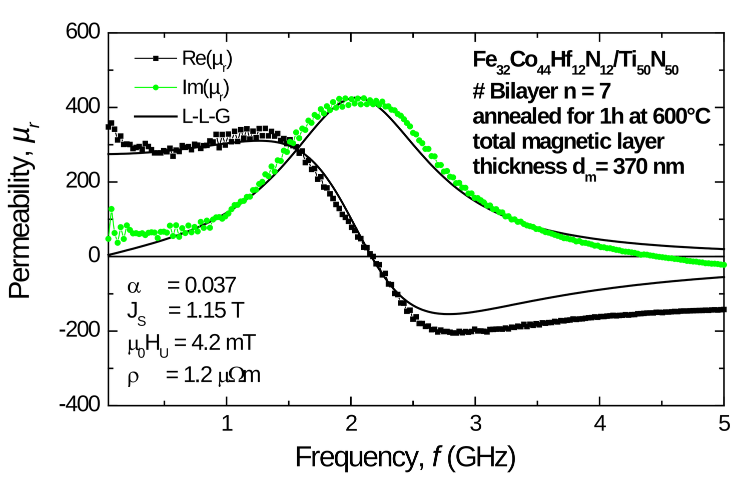
➢ **$Fe_{32}Co_{44}Hf_{12}N_{12}/Ti_{50}N_{50}$ multilayer films annealed at T_a = 600 °C for 1 h are suitable for detecting changes in the resonance frequency up to 500 °C**

Kittel resonance formula: $f_r = \frac{\gamma}{2\cdot\pi}\cdot\sqrt{(\mu_0 H_u)^2 + J_S\cdot\mu_0 H_u}$

- Decrease in J_s and μ_0H_u

➢ f_r decreases with increasing temperature

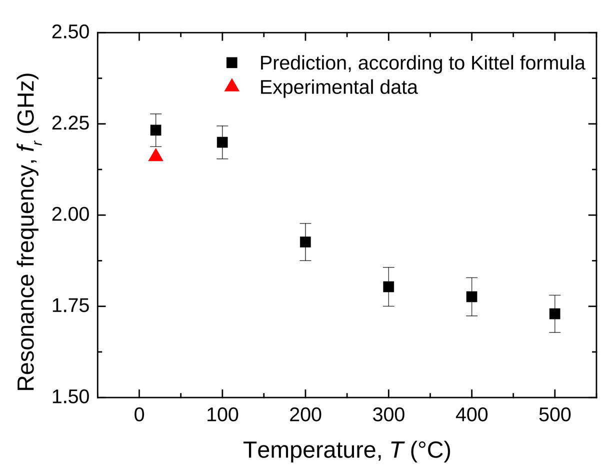
Experiment: f_r at 20 °C



Dynamic behavior of magnetic moments in a HF-field: Landau-Lifschitz-Gilbert equation (L-L-G) [1] in combination with the Maxwell equations to consider eddy-currents [2]:

$$\frac{\partial \boldsymbol{M}}{\partial t} = -\gamma \boldsymbol{M} \times \boldsymbol{H}_{eff} + \frac{\alpha_{eff}}{M_S}\left(\boldsymbol{M} \times \frac{\partial \boldsymbol{M}}{\partial t}\right)$$

Prediction: f_r with increasing temperature

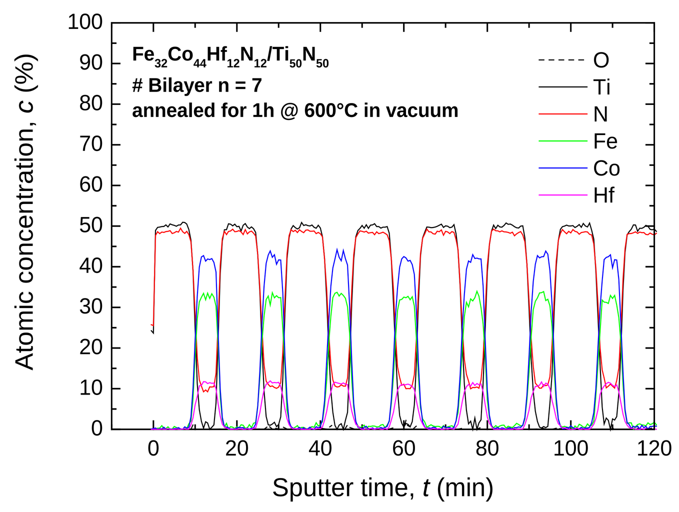


- Kittel formula: a decrease in f_r is predicted due to the decrease in J_s(T) and μ_0H_u(T) with increasing temperature
- 20 °C: f_r was confirmed experimentally
- Due to thermal fluctuations the damping parameter α is expected to increase
- f_r(T) will also be affected by α(T)

Oxidation process due to heating in air?

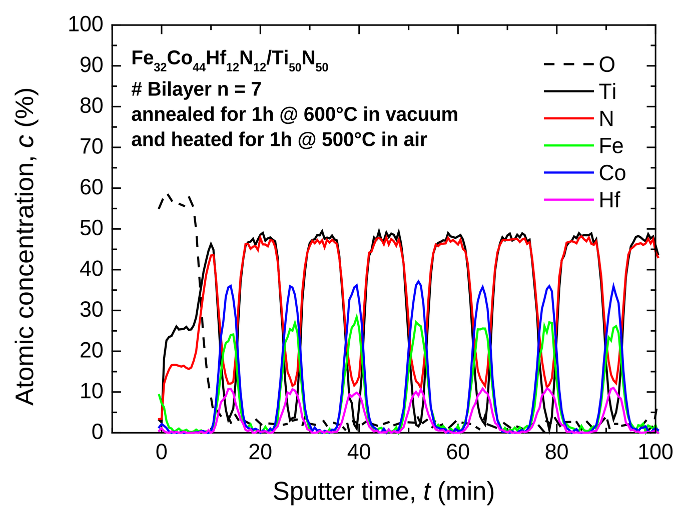
Auger electron spectroscopy depth profiles

Before heating up to 500 °C:



- No oxygen in the multilayer film due to annealing in vacuum

After heating up to 500 °C in air for 1 h during the measurement :

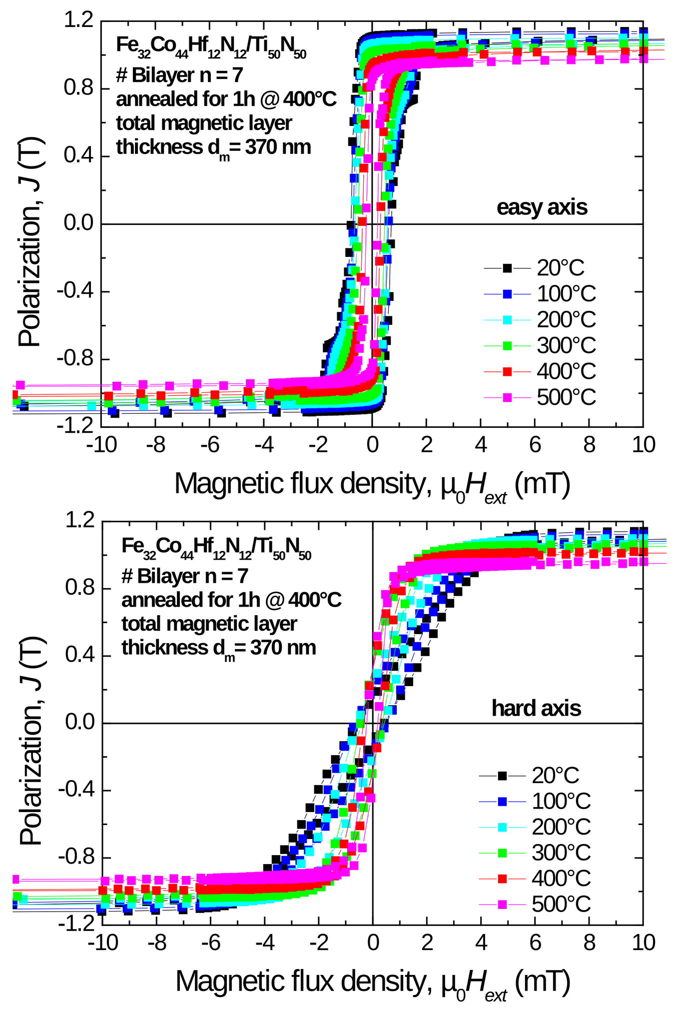


- TiN top layer has oxidized to a large extent to TiO_2
- A diffusion of the oxygen to the magnetic $Fe_{32}Co_{44}Hf_{12}N_{12}$ layer has not occurred

➢ **Ferromagnetic properties are maintained**

Multilayer films annealed for one hour at T_a = 400 °C in vacuum

Temperature-dependent hysteresis loop measurements in easy and hard axis of polarization from RT up to 500 °C in air



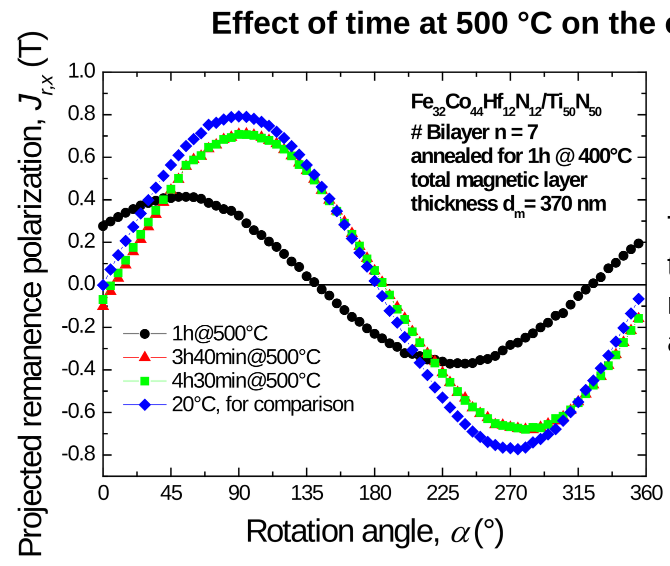
- Clear distinction between easy and hard axis at RT
- Above 200 °C the clear distinction starts to vanish

➢ The direction of μ_0H_u seems to shift out of its originally preferred direction

➢ Hysteresis loop measured in the "hard axis" of polarization shows a decreasing uniaxial anisotropy field μ_0H_u

➢ **$Fe_{32}Co_{44}Hf_{12}N_{12}$ / $Ti_{50}N_{50}$ multilayer films annealed at T_a = 400 °C for 1 h are less suitable for detecting changes in the resonance frequency above 200 °C**

Effect of time at 500 °C on the orientation of μ_0H_u:



The direction of μ_0H_u relaxes towards its originally direction at room temperature after 3 h 40 min at 500 °C

Summary

- By annealing the $Fe_{32}Co_{44}Hf_{12}N_{12}/Ti_{50}N_{50}$ multilayer films at either T_a = 400 °C or 600 °C for 1 h in a static magnetic field in vacuum a uniaxial anisotropy field of about μ_0H_u ≈ 5 mT was induced
- The films annealed at T_a = 600 °C show a temperature stability of μ_0H_u up to 500 °C at least for 1 h

➢ **Thermally induced strain relaxes instantaneously**

➢ **$Fe_{32}Co_{44}Hf_{12}N_{12}$ / $Ti_{50}N_{50}$ multilayer films annealed at T_a = 600 °C for 1 h are suitable for detecting changes in the resonance frequency up to 500 °C**

- In contrast, the films annealed at T_a = 400 °C lose this metastable state above 200 °C, because the orientation of μ_0H_u in the film plane has shifted out of its room temperature direction
- The change of the uniaxial anisotropy field direction could have been caused by mechanically and thermally induced strain in the magnetostrictive material

➢ **Thermally induced strain starts to relax after approximately 3 h at 500 °C**

➢ **$Fe_{32}Co_{44}Hf_{12}N_{12}/Ti_{50}N_{50}$ multilayer films annealed at T_a = 400 °C for 1 h are less suitable for detecting changes in the resonance frequency above 200 °C**

Outlook

Uniaxial anisotropy field:

- Temperature stability of μ_0H_u depends on a possible oxidation process of the magnetic layer

➢ Further investigations on the oxidation process at high temperatures

Temperature dependent resonance frequency:

- Verification of the thermal stress
- Integration of the thermally induced residual stress in the model for f_r(T) by introducing a magnetoelastic anisotropy
- Experimental verification of f_r(T)

[1] T.L. Gilbert, IEEE Trans. Magn. 40 (2004)
[2] K. Seemann, H. Leiste, V. Bekker, J. Magn. Magn. Mater. 278 (2004)