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Combined ferromagnetic and mechanical properties of $Fe_{32}Co_{44}Hf_{12}N_{12}$/TiN multilayer coatings for high frequency sensor applications

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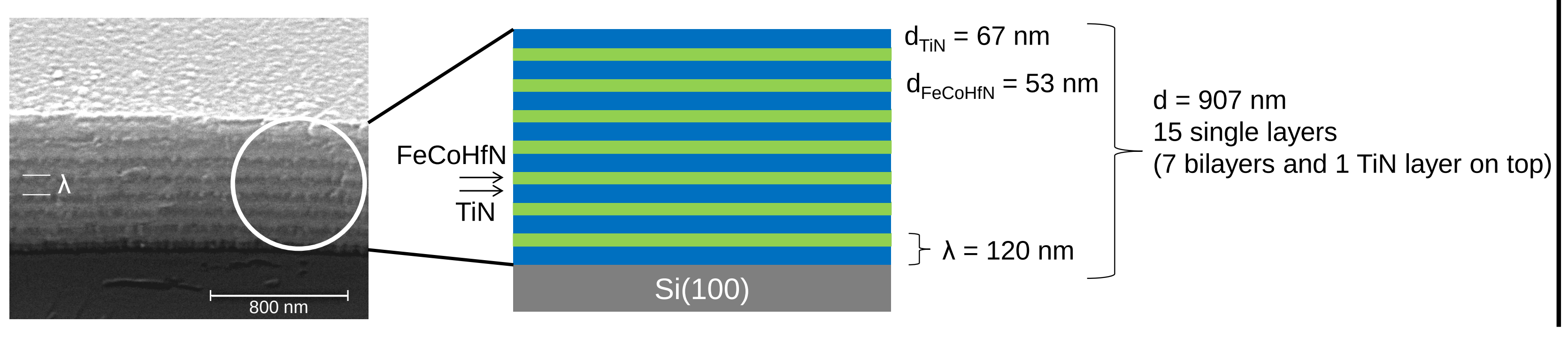
Nowadays, as the state of the art, wear resistant coatings are used to extend the lifetime of mechanical components like, e.g., bearings or tools.
As a new concept, we combine hardness with magnetic properties within a thin film multilayer system which consists of hard and ferromagnetic interface layers, in order to simultaneously develop the mechanical and magnetic properties as a functional sensor material.
This multilayer acts as a wear resistant coating, and due to the embedded ferromagnetic material it is possible to monitor an in situ temperature or strain induced stress which changes the magnetic properties like high frequency response.
We present the investigations of a new FeCoHfN/TiN multilayer system with regard to its hardness and ferromagnetic high frequency properties in comparison to FeCoHfN and TiN single layers, in order to obtain an appropriate sensor material.

Sample preparation:

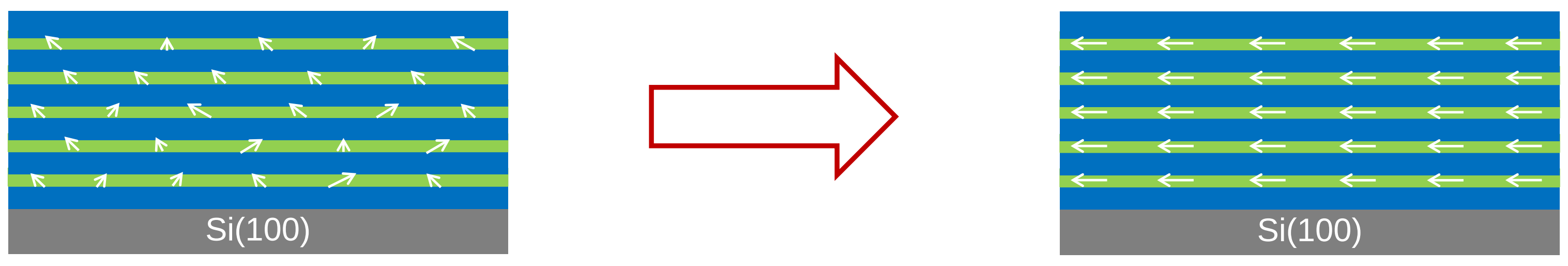
- Reactive dc and rf magnetron sputtering from a $Fe_{37}Co_{46}Hf_{17}$ and a $Ti_{50}N_{50}$ target in Ar and N_2 atmosphere with $N_2/Ar = 3\%$ and p = 0.2 Pa
- Alternating deposition of TiN and FeCoHfN single layers with modulation period λ = 120 nm



Heat treatment:

- Annealing for 1h at 600°C in a static magnetic field (50 mT) in vacuum after deposition
  - ➢ Inducing a uniaxial anisotropy H_U for a homogenous precession of magnetic moments in an external field

Annealing in a static magnetic field



Requirements for high frequency sensor applications:

- Resonance frequency in the GHz regime
  - ➢ Exact Kittel resonance formula for uniformly magnetized samples:

$$f_r = \frac{\gamma}{2 \cdot \pi} \cdot \mu_0 \cdot \sqrt{H_U^2 + M_S \cdot H_U}$$

- High initial permeability at low frequencies:

$$\mu_r'(f \to 0) = \frac{J_S}{\mu_0 H_U} + 1$$

- Thermal stability at high temperatures

- High uniaxial anisotropy H_U
- High saturation polarization J_S
- Low coercive field H_C
  - ➢ $H_C \sim \frac{K_1^4 d^6}{J_S A^3}$ [1] ⟶ low magnetocrystalline anisotropy $K_1 \approx 0$ or d small for soft magnetic materials
  - ➢ Refractory metal-nitride grain boundaries of HfN acting as grain reducer

FeCo
✚ HfN

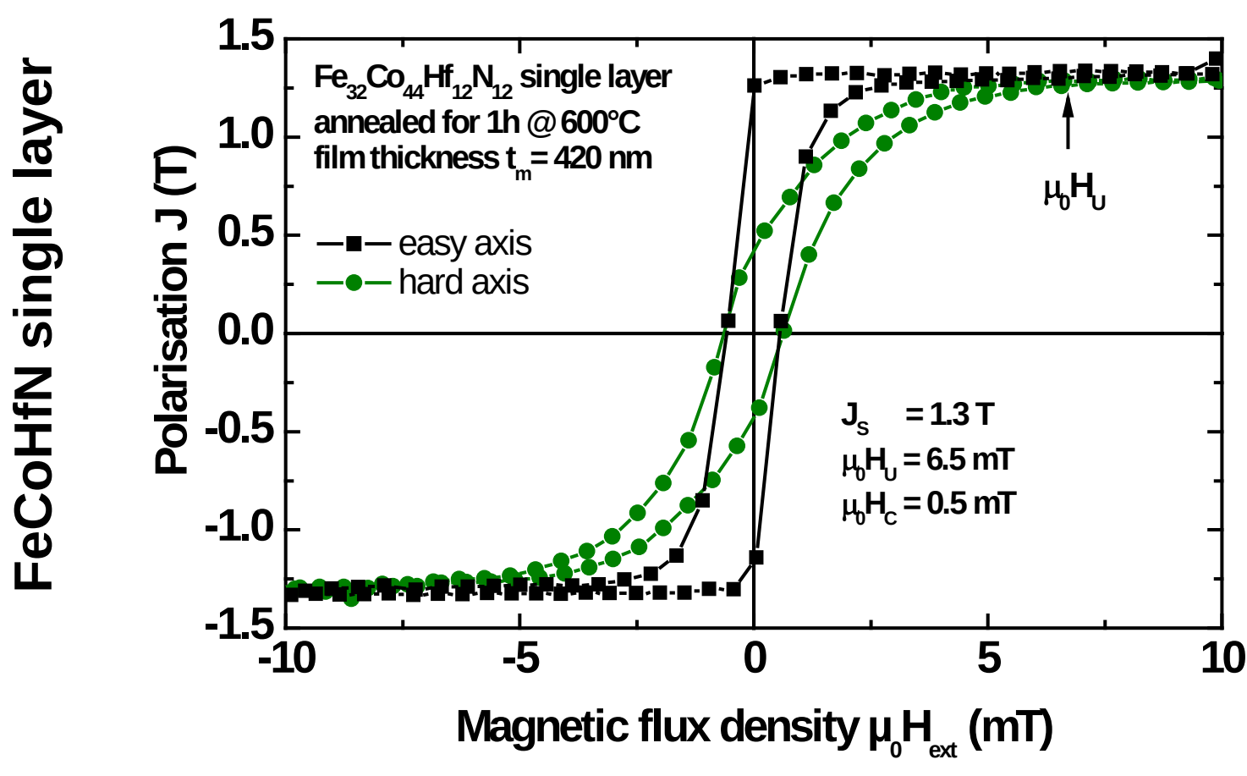
Ferromagnetic material **Wear resistant coating**

FeCoHfN ✚ TiN

Functional sensor material

Ferromagnetic properties

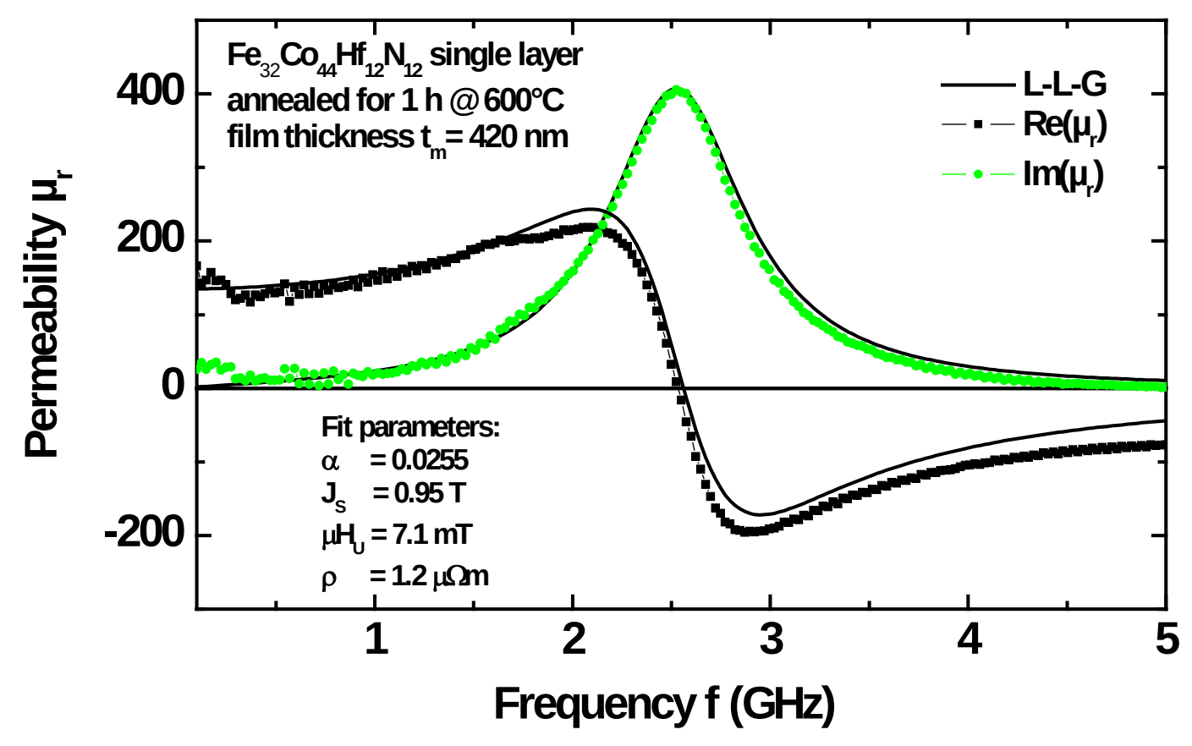
Vibrating Sample Magnetometer



- High saturation polarization
- Low coercive field
- ➢ Soft magnetic properties due to annealing in a static magnetic field
- ➢ Distinction between easy and hard axis as a result of successfully induced unidirectional in-plane anisotropy possible

- ➢ But due to annealing also magnetocrystalline anisotropy increases and grain growth
- ➢ A marked uniaxial anisotropy field cannot be identified

Frequency dependent permeability



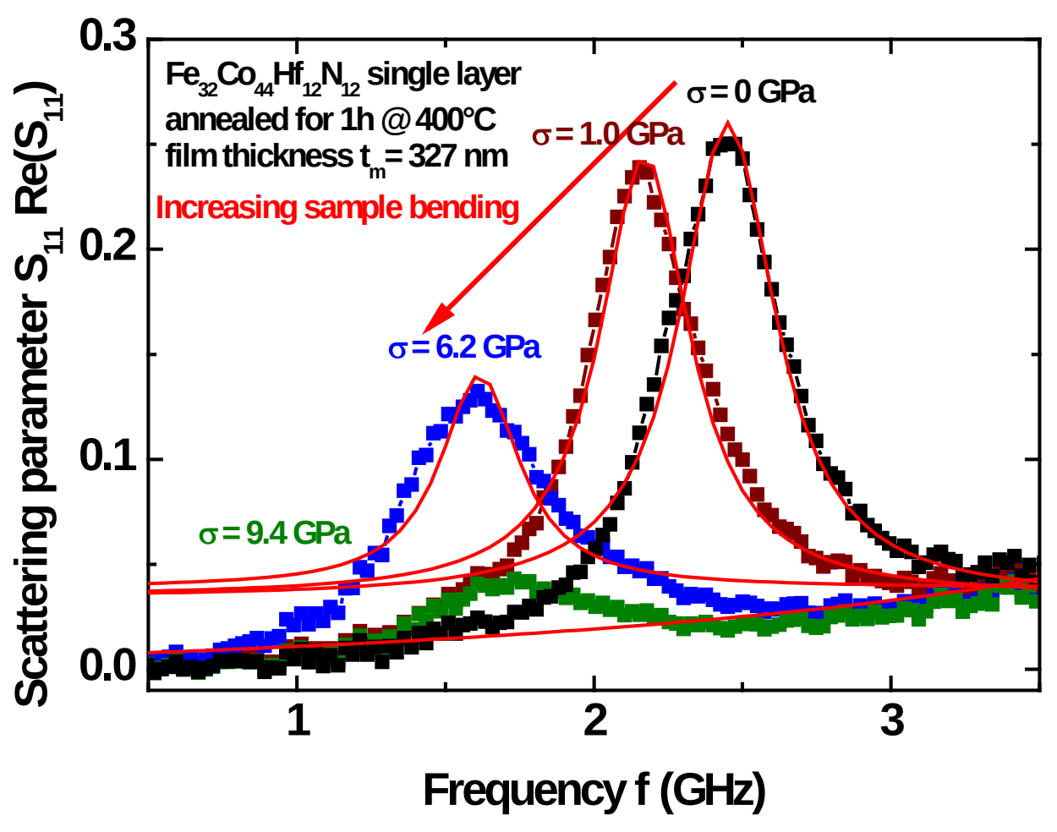
- Dynamic behavior of magnetic moments exposed to an electromagnetic wave: Landau-Lifschitz-Gilbert equation (LLG) in combination with the Maxwell equations to consider eddy-currents:

$$\frac{\partial \boldsymbol{M}}{\partial t} = -\gamma \boldsymbol{M} \times \boldsymbol{H}_{eff} + \frac{\alpha_{eff}}{M_S}\left(\boldsymbol{M} \times \frac{\partial \boldsymbol{M}}{\partial t}\right) \text{[2]}$$

- High resonance frequency of f_r = 2.56 GHz

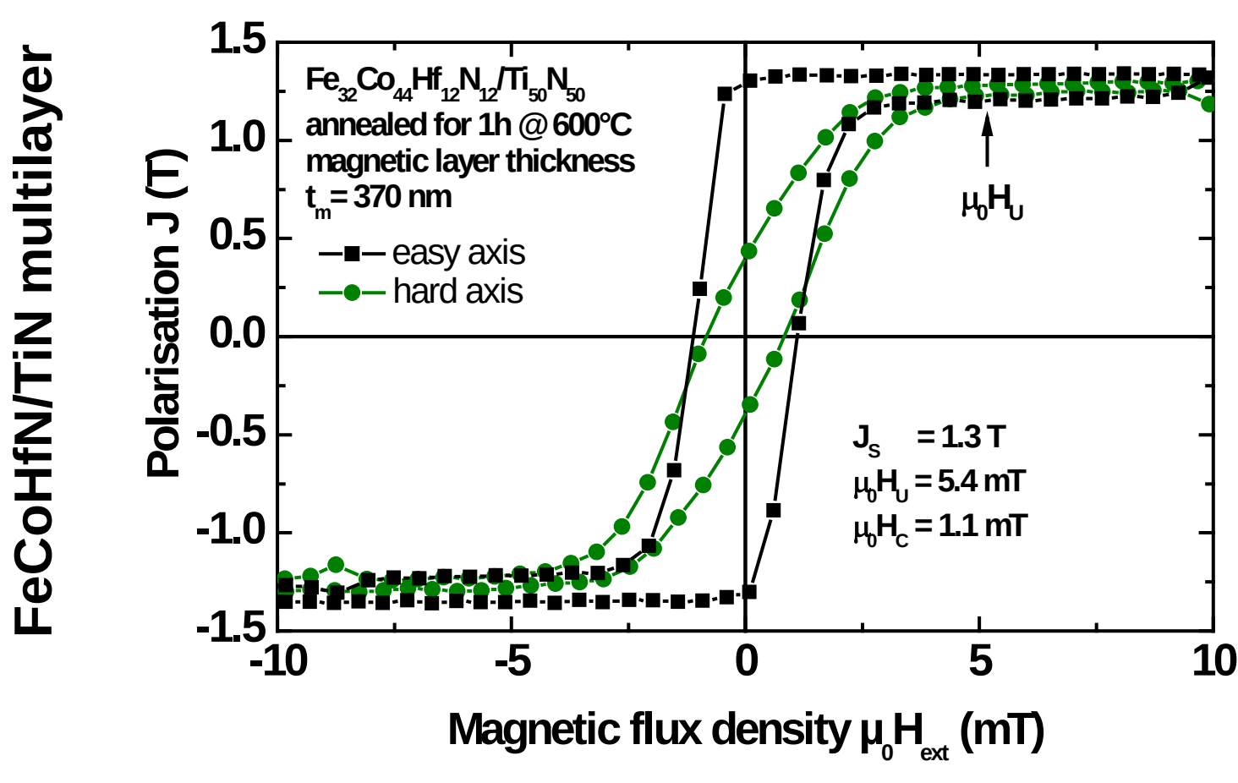
- Initial permeability at low frequency μ_r' = 135 is relatively low
- ➢ Orientation of magnetic moments in one direction constrained by magnetocrystalline anisotropy
- ➢ Relatively high damping because of HfN phase which acts as scattering center

Magneto-elastic measurements

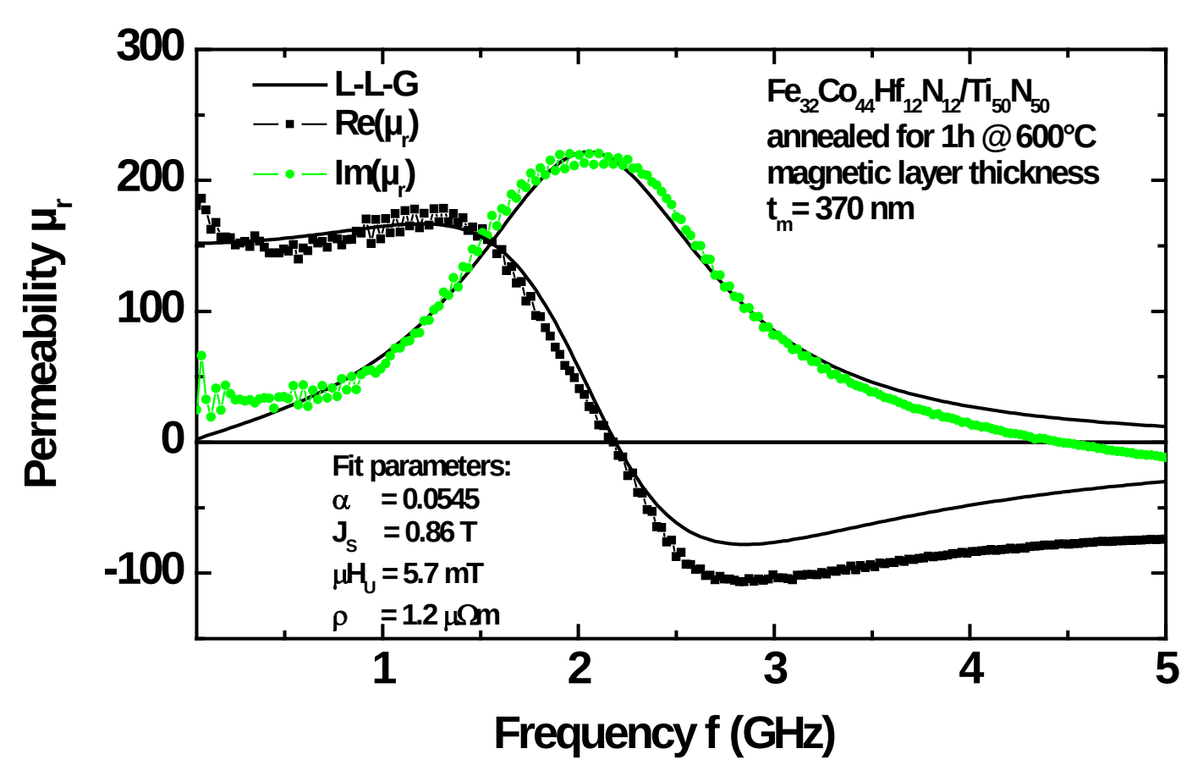


- Shift from f_r = 2.45 GHz (σ = 0 GPa) to f_r = 1.62 GHz (σ = 6.2 GPa) for single film (from Lorentz fits)

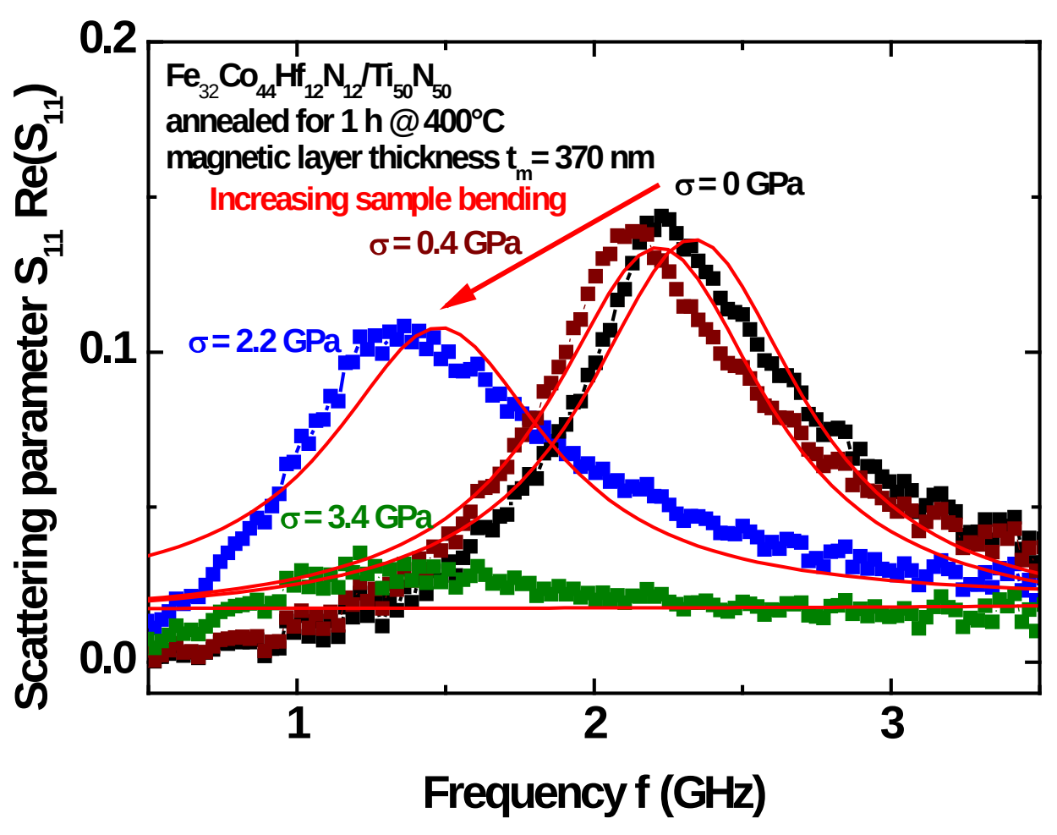
- High frequency sensor applications:
  - ➢ Detecting changes in resonance frequency caused by magneto-elastic effects due to inverse magnetostriction (Villari effect)
  - ➢ Mechanical stress perpendicular to the preferred magnetic direction impacts the magnetic anisotropy
  - ➢ Generates rotation of the magnetic moments which shifts the ferromagnetic resonance frequency



- Saturation polarization of magnetic material same as for single film
- Uniaxial anisotropy less than for single film
- Coercive field greater than for single film
- ➢ Interface layers and ordering processes influence ferromagnetic properties
- ➢ But soft magnetic properties are still conserved and a uniaxial anisotropy could be induced



- Additional TiN and interface layers affect the high frequency response
- ➢ Higher damping as for the single film due to higher fraction of nonmagnetic material
- ➢ Effective saturation polarization much smaller
- ➢ Resonance frequency f_r = 2.18 GHz less than for single film but still above 2 GHz
- ➢ Initial permeability at low frequency μ_r' = 152 is greater than for the single film



- Shift from f_r = 2.33 GHz (σ = 0 GPa) to f_r = 1.48 GHz (σ = 2.2 GPa) for multilayer system (from Lorentz fits)
- ➢ Nearly a change of 1 GHz in resonance frequency
- ➢ Detectable sensor signal

Mechanical properties

Residual stresses

Material	Treatment after deposition	Film thickness (µm)	σ (GPa)
$Ti_{50}N_{50}$	as deposited	2.046	0.34 ± 0.10
$Ti_{50}N_{50}$	1h@600°C	2.046	0.43 ± 0.06
$Fe_{32}Co_{44}Hf_{12}N_{12}$	as deposited	0.420	-1.11 ± 0.08
$Fe_{32}Co_{44}Hf_{12}N_{12}$	1h@600°C	0.420	1.45 ± 0.17
$Fe_{32}Co_{44}Hf_{12}N_{12}/Ti_{50}N_{50}$	as deposited	0.904	-2.66 ± 0.09
$Fe_{32}Co_{44}Hf_{12}N_{12}/Ti_{50}N_{50}$	1h@600°C	0.904	0.13 ± 0.15

- Compressive stress due to growth conditions during the sputtering process for FeCoHfN single layer
- Large mismatch between lattice constants of TiN and FeCo results in very high compressive stress for the as deposited FeCoHfN/TiN multilayer
- After annealing stress becomes tensile because of different thermal expansion coefficients of layers and substrate
- Lowest stress for annealed multilayer

Hardness and reduced Young´s modulus

Material	Treatment after deposition	Film thickness (µm)	H (GPa)	E_r (GPa)
$Ti_{50}N_{50}$	as deposited	1.548	18.5 ± 1.4	205.0 ± 5.3
$Fe_{32}Co_{44}Hf_{12}N_{12}$	as deposited	0.420	10.8 ± 3.5	119.6 ± 3.3
$Fe_{32}Co_{44}Hf_{12}N_{12}/Ti_{50}N_{50}$	as deposited	0.904	14.6 ± 1.9	160.1 ± 4.6
$Fe_{32}Co_{44}Hf_{12}N_{12}/Ti_{50}N_{50}$	1h@600°C	0.904	17.2 ± 1.2	175.1 ± 4.0

- Hardness of as deposited multilayer is in good agreement with the rule of mixture value (H = 15.3 GPa)
- As a result of annealing the hardness of the multilayer approaches nearly the value of TiN

Conclusions

$Fe_{32}Co_{44}Hf_{12}N_{12}$/TiN multilayer coating:

- Resonance frequency above 2 GHz
- Stress induced changes in high frequency response could be measured
- Mechanical hardness of the annealed multilayer near the value of TiN
- Very small residual stresses after annealing

Hardness and ferromagnetic high frequency properties could be combined in one functional $Fe_{32}Co_{44}Hf_{12}N_{12}$/TiN material system

References

[1] G. Herzer, IEEE Trans. Magn. 26 (1990) 1397
[2] K. Seemann et al., J. Magn. Magn. Mater. 302 (2006)