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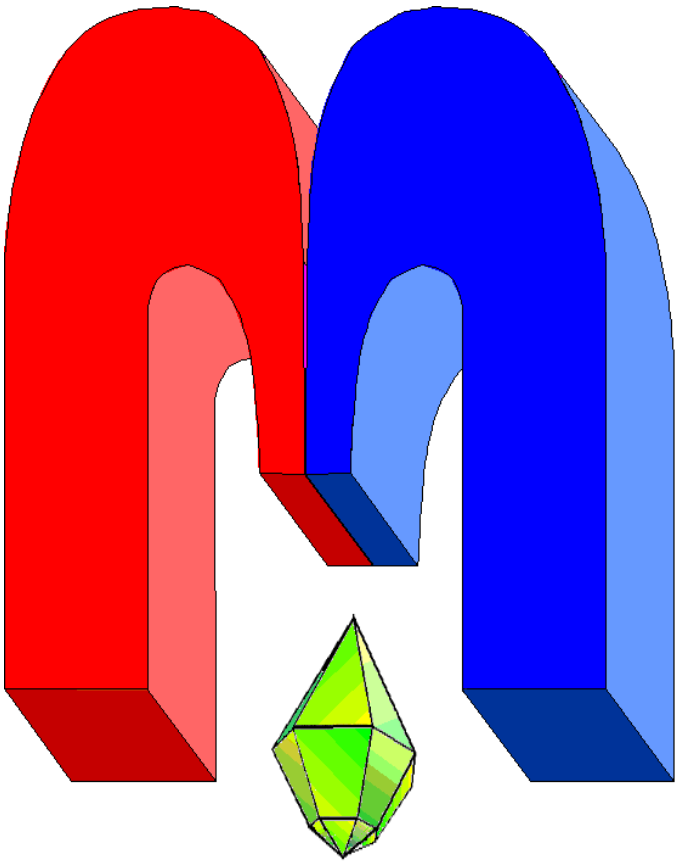

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* In Kazan University the Electron Paramagnetic Resonance (EPR) was discovered by Zavoisky E.K. in 1944.

Magnetic and magnetoresonant properties of thin-film Fe/Ag/Co/CoO heterostructure synthesized by combined molecular beam epitaxy and reactive magnetron sputtering

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In the paper, we report on growth conditions, structure characterization, magnetic and magnetoresonance studies of thin-film Fe/Ag/Co/CoO heterostructure deposited ontop of the single-crystal (001)-MgO substrate utilizing *in situ* combination of molecular beam epitaxy and magnetron sputtering. The molecular beam deposition was applied to grow the first, iron layer epitaxially on MgO realizing the 45-degree lattice matching between the magnesium oxide and the body-centered cubic iron. Subsequent silver layer was also grown epitaxially with the favorable 45-degree lattice matching between the iron and silver. Next, Co-metal and the top, cobalt oxide, layers were deposited by magnetron sputtering and reactive sputtering, respectively, after moving the sample into a magnetron sputtering chamber without breaking the ultra-high vacuum conditions. Magnetometry and ferromagnetic resonance studies of the resulting heterostructure have revealed a combination of the four-fold and uniaxial anisotropies in the film plane at temperatures well above the Neel temperature for antiferromagnetic CoO ($T_N = 291$ K). After field-cooling of the sample down to $T = 180$ K, ferromagnetic resonance measurements following the major magnetic hysteresis loop have shown a correlated precession of the iron and cobalt layers magnetizations giving the evidence of the interlayer coupling through the silver interlayer.

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Keywords: magnetic anisotropy, ferromagnetic resonance, magnetron sputtering, magnetic hysteresis, interlayer coupling.

1. Introduction

The interlayer coupling between ferromagetic layers through a non-magnetic spacer in ultrathin heterostructures, discovered in 1988 by A. Fert [1] and P. Gruenberg [2], was put into a base of modern spintronics [3]. The topic was intensely developed in mid-90th and finally was implemented in high-performance hard disk drive (HDD) heads providing a fantastic increase in their capacity (from 2-4 GB for the coil heads at mid-90th to 14 TB of nowadays data-server HDDs utilizing shinglass recording technology; see, for example, Refs. [4, 5] and citations therein). The revival of interest to the interlayer coupling in the last decade was associated with the studies of the spin moment transfer between ferromagnetic layers in magnetic heterostructures. This idea and the physics of spin-transfer torque were implemented in magnetic random access memory devices promising sub-nanosecond access time, non-volatility and mechanical robustness (see, for example, Refs. [5, 6] and citations therein). Recent proposal of an ultrafast magnetic switching at optical frequencies [7] has triggered new interest to multilayer heterostructures combining different magnetic materials. Our study deals with the thin-film Fe/Ag/Co/CoO heterostructure comprising interacting layers of iron and cobalt and showing coupled dynamics at frequencies of tens of gigahertz.

2. Sample preparation

A series of Fe/Ag/Co/CoO heterostructure samples was prepared by sequential deposition of the elements from the molecular beam sources using ultra-high vacuum (UHV) molecular beam epitaxy (MBE) chamber (*SPECS GmbH*, *Germany*) and UHV magnetron sputtering (MS) chamber (*BESTEC*,

Germany) of a combined UHV vacuum system. The base vacuum in the MBE chamber was better than 3×10^{-10} mbar before filling-in the liquid nitrogen jacket, and better than 8×10^{-11} mbar after the jacket cooling. The residual gas pressure in the MS chamber was better than 5×10^{-9} mbar. The UHV transfer line allowed us to move samples from one chamber to another without breaking the UHV conditions. 5D sample holder stage, equipped with an *e*-beam powered heater, allows to keep the substrate at a temperature of up to 1150°C upon annealing or deposition. The growth rate was monitored in a real time using a calibrated quartz-crystal oscillator (by *INFICON*, *Switzerland*) with an accuracy of 10^{-3} nm/sec and a capability of calculating an actual film thickness. The *BRUKER Dektak XT* stylus profiler (the stylus tip radius is 2 μm, step height measurement accuracy is better than 0.5 nm) was used to measure *ex situ* the resulting thickness of the samples utilizing the shadow mask method. Crystallinity of the films could be inspected *in situ* by low-energy electron diffraction (LEED by *SPECS*) technique at any desired step of the deposition process. The elemental compositions were verified by X-ray photoelectron spectroscopy (XPS *by SPECS*) setup built-in the analytical chamber (ACh). XPS measurements were performed without breaking the vacuum since the MBE and ACh are connected with an UHV transfer line. The analytical chamber operates at the base pressure of $\sim 3\times10^{-10}$ mbar; it is equipped with the Mg-K_α X-ray source operating at 12.5 kV and 250 W, and the *Phoibos* 150 hemispherical energy analyzer of photoelectrons (all from *SPECS*). The survey XPS spectra were recorded in the range of 0-1000 eV with the energy step of 1 eV and the pass energy of 80 eV. High-resolution spectra were recorded in limited energy ranges of interest by averaging over 100 scans for each element with the energy step of 0.1 eV and the pass energy of 20 eV.

We have chosen single-crystal, (001)-oriented epi-polished MgO plates (*Crystal GmbH*, *Germany*) as a substrate for our multilayer. The choice of the substrate was ruled by the good matching of the lattice constants and cubic symmetry of the magnesium oxide ($a_{MgO} = 421.6$ pm) and iron ($a_{Fe} = 285.6$ pm; $a_{Fe}\sqrt{2} = 403.9$ pm). Then, these parameters provide the conditions for 45-degree epitaxial growth of the iron film directly on the MgO (001) substrate because of a small mismatch of about 4.5% between the lattices. The MgO substrates were first thoroughly rinsed in several steps using ultrasound bath and subsequently annealed in vacuum at 800°C for 5 minutes in the molecular-beam epitaxy (MBE) chamber to remove the residual surface contaminants. Then, metallic iron (4N purity) was evaporated from an effusion cell at a rate of 1Å/min onto the (100)-MgO substrate kept at the temperature of 100ºC. The layer thickness was varied in the range of 1-5 nm. Subsequently, a 4 nm thick layer of cubic symmetry silver (4N purity, $a_{Ag} = 408.5$ pm) was epitaxially grown with the 45-degree matching on top of the Fe-layer by evaporation from another effusion cell at a rate of 1Å/min.

To deposit metallic cobalt and insulating antiferromagnetic cobalt oxide layers, the sample holder with the substrate and the deposited two layers was moved from the MBE chamber to a magnetron sputtering (MS) chamber without breaking the UHV conditions. The base pressure in the UHV MS chamber of about 5×10^{-9} mbar prevented any degradation of the silver film during several minutes of the transfer procedure. Then, in the MS chamber, a cobalt layer was deposited with the substrate, kept at room temperature, by sputtering a metallic cobalt target (4N purity) to complete the core of the heterostructure. For the magnetron operation, an ultra-pure argon (6N purity) was used as a plasma gas at a pressure of 6×10^{-3} mbar. Finally, the antiferromagnetic cobalt oxide layer was reactively deposited by sputtering the metallic cobalt target in a plasma of calibrated Ar and O_2 gas mixture. This layer is necessary to exchange bias the adjacent Co layer to make it magnetically hard. A composition of the CoO layer was examined by XPS to verify the stoichiometry of the oxide. It is very important because several cobalt oxides exist, from which Co_3O_4 has low Neel temperature of $T_N = 40$ K, and only CoO has $T_N = 291$ K and is suitable for our purposes. The XPS spectrum has shown the Co : O = 1 : 1 composition of our oxide (see Fig. 1a). After finishing the sample preparation it was retrieved from the vacuum system for *ex situ* magnetic measurements.

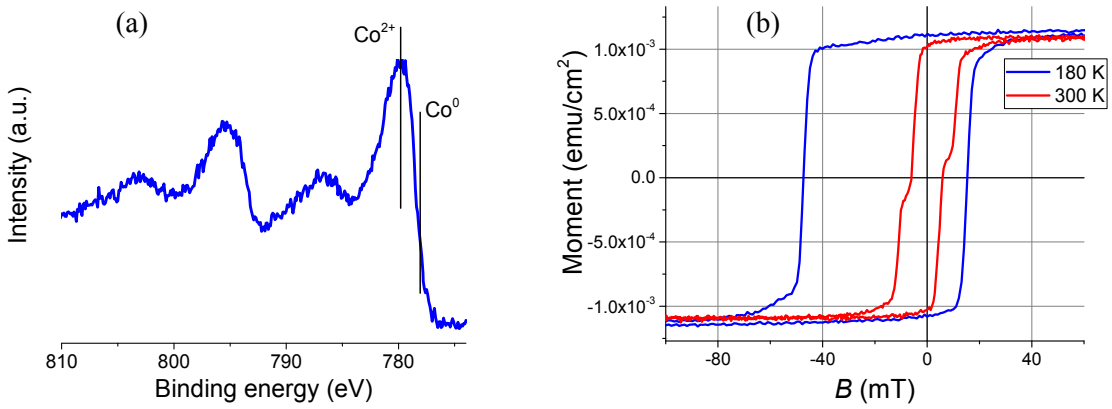

Figure 1. X-ray photoemission spectrum of the Fe/Ag/Co/CoO heterostructure on (001)-oriented MgO in the range of the energies of 2p-electrons of cobalt (a); magnetic hysteresis curves of the heterosctructure measured at $T = 300$ K and at $T = 180$ K after cooling in the magnetic field of 400 mT applied along the [110] direction of the MgO substrate (b).

3. Magnetometry and ferromagnetic resonance measurements and their analysis

The resulting Fe/Ag/Co/CoO heterostructures were characterized by measuring the magnetic hysteresis with the *Quantum Design PPMS-9* vibrating sample magnetometer (VSM) setup (the temperature range 2-400 K and the magnetic field of up to ±9 T). The magnetic field was applied parallel to the film plane along the [110] direction of the MgO substrate. The sample was cooled from room temperature (RT) to a measurement temperature in a field of 400 mT. Strong exchange bias (leftward shift of the magnetic hysteresis loop) was observed after field-cooling of the samples to 180 K (see Fig. 1b), and the saturation magnetizations of the layers were determined from these measurements.

Further, the magnetic anisotropies of the structure were studied by the ferromagntic resonance technique (FMR) with the X-band *Bruker ESP300cw* electron spin resonance (ESR) spectrometer equipped with the *Oxford Instruments* ER4111VT nitrogen-flow system operating in the temperature range of 90-550 K and magnetic fields up to 1.313 MA/m (16.5 kOe). The standard ER4116DM high-Q double-mode rectangular TE_{102}-mode cavity was used in the measurements. The FMR data were first recorded at the elevated temperature of $T = 355$ K, well above the Neel temperature of CoO $T_N = 291$ K, to avoid any magnetic fluctuations influence from this layer. The measured spectra are presented in Fig. 2a. They show clear angular variation, and the dependence of the resonance field

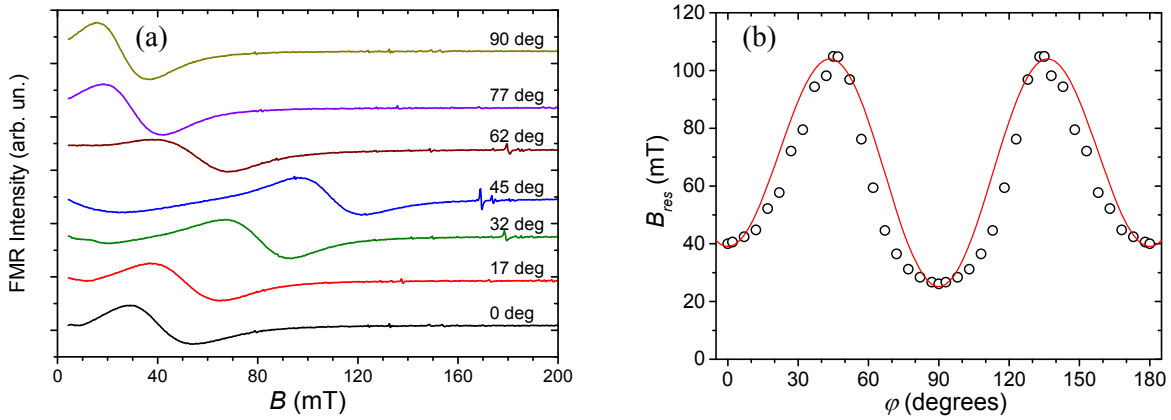

Figure 2. In-plane angular variation of the ferromagnetic resonance spectrum of the Fe/Ag/Co/CoO heterostructure on MgO (001) at $T = 355$ K (a); angle $\varphi = 0$ degrees corresponds to the [110] crystallographic direction of the MgO substrate. In-plane orientation dependence of the resonance field value (b).

extracted by fitting the FMR spectra is presented in Fig. 2b. The angular dependence can be fairly well described by a superposition of the four-fold and two-fold anisotropies (the simple model result is drawn by the red solid line). Since the iron layer is epitaxial and grown on high-quality (001)-oriented MgO, it must possess the four-fold anisotropy [8]. The silver layer ontop the iron should not itself introduce lattice distortion because of the cubic symmetry and extremely good lattice matching with iron (a_{Ag} = 408.5 pm against 403.9 pm for the epitaxial 45-degree mode of the growth). Then, we may think that the cobalt layer could be a source of the uniaxial anisotropy. Indeed, it is known that ultrathin Co-films grown on gold (properties very similar to silver) reveal the uniaxial anisotropy [9].

Now, the question arises how the uniaxial cobalt influences the resonance properties of the iron layer. Here, the lattice distortion transferred through the silver layer can in principle be considered. However, it is hardly likely that 4 nm of cobalt can transfer its possible uniaxial distortion through 4 nm of mechanically-soft metallic silver to a much more mechanically hard iron by elastic compression-expansion. It is worthy to mention that we observe a single pronounced FMR line with the angular dependence contradicting the FMR findings of Ref. [9]. The observed FMR line certainly represents a synphase ("acoustic") mode of the coupled precession of the two, iron and cobalt, layers, which may occur only if the layers are interacting.

To shed light on the situation we have performed additional FMR measurements at 180 K well below the CoO Neel temperature (see Fig. 3). The sample was field-cooled to 180 K in the field of 400 mT applied along the easy axis direction of the FMR signal in Fig. 2b (φ = 90 degrees). Then, the measurement has been performed following the magnetic hysteresis loop in Fig. 1b (blue solid line) utilizing the following procedure. Step 1: the magnetic field was decreased from 400 mT to 3 mT with a synchronous recording of the FMR spectrum. Because of a single-polarity electromagnet of the Bruker spectrometer, at zero field the sample was rotated by 180 degrees to continue following the

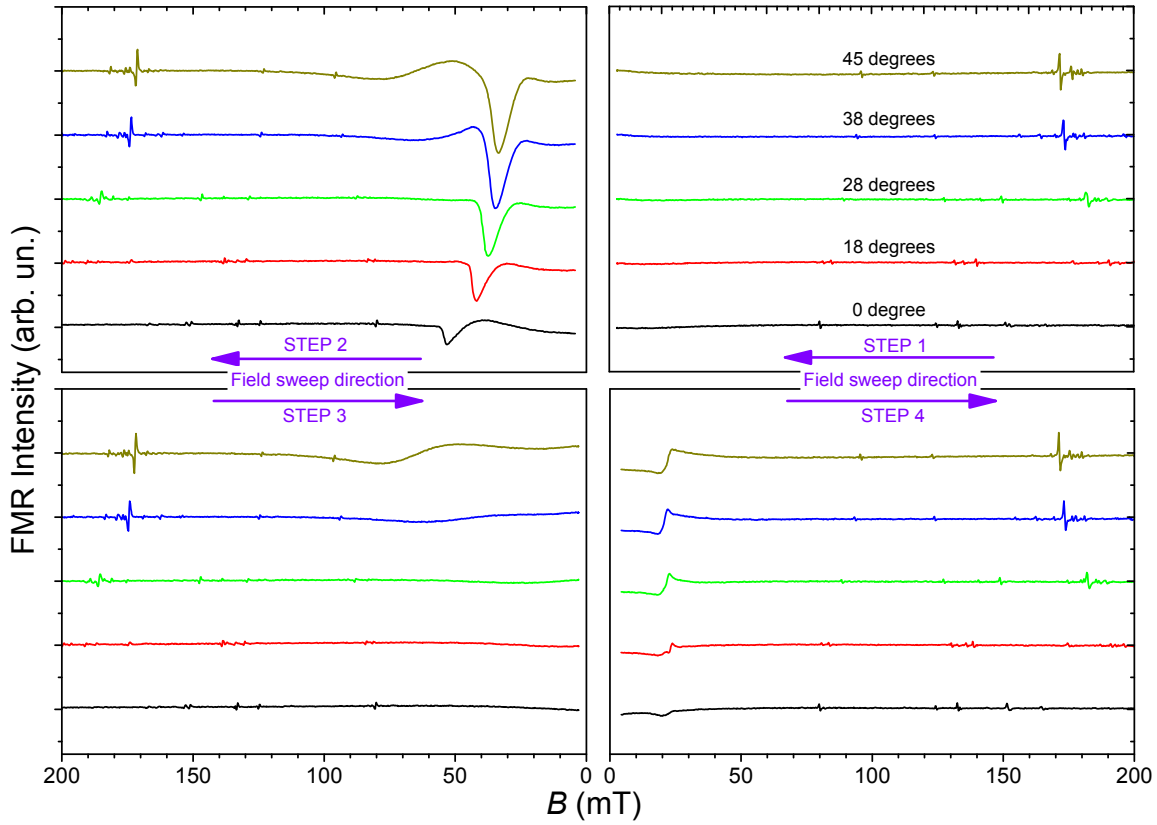

Figure 3. Ferromagnetic resonance spectra of the Fe/Ag/Co/CoO heterostructure on (001) MgO measured in a cyclic manner at T = 180 K after cooling in the magnetic field of 400 mT applied along the [110] direction of the MgO substrate. Angles between the magnetic field $\mathbf{B}_0$ and [110] direction of MgO are indicated in the upper right panel.

hysteresis loop applying the positive-directed magnetic field. Step 2: the field was increased mimicking negative-directed one in Fig. 1b, and the FMR spectrum was continuously recorded upon the field sweep. At 400 mT the sweep was stopped and (Step 3) the field again was decreased to 3 mT with the FMR spectrum continuously recorded. At $B = 3$ mT, the sample was again 180 degrees rotated, and (Step 4) the field was subsequently increased to 400 mT to the initial cooling-field value. The FMR spectra collected with this way are presented in Fig. 3 for different angles between the external field upon the FMR spectrum collection, and the direction of the cooling field (exchange bias direction). The FMR spectra of Fig. 3 reveal a clear irreversibility introduced by a presence of the exchange bias. When recorded at small angles with respect to the cooling field direction (see Fig. 3) the FMR spectra show abrupt inflection occurring at the coercive field of the magnetic moment reversal of the heterostructure (see Fig. 1b). In general, the FMR spectra in Fig. 3 show presence of two resonance signals visible due to strong internal field induced by the exchange bias. Then, the fact that the second signal is not accessible in the weak magnetic fields covered in the spectra in Fig. 2a, give evidence that indeed we observe there the acoustic precession mode of the coupled Fe/Ag/Co bilayer.

4. Summary

The thin-film Fe/Ag/Co/CoO heterostructures were grown on single-crystal (001)-oriented MgO substrate combining molecular beam epitaxy and reactive magnetron sputtering techniques. The VSM magnetometry measurements have shown that the field-cooled samples acquire pronounced exchange bias for the field applied in the structure plane. Subsequent ferromagnetic resonance measurements at temperatures above and below the Neel temperature of cobalt oxide have shown correlated precession of the layers magnetizations giving evidence of interlayer coupling between the iron and cobalt films through the silver interlayer.

Acknowledgments

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