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Abstract. Magnetic, resonance and transport properties of Fe(t nm)/MgO(3.0 nm) multilayers prepared by pulsed laser deposition were investigated. Comparison of the data allows conclusions on Fe layers morphology. For t<0.61 nm typical features of granular cermet films in dielectric regime are observed, i.e. high electrical resistance, isotropic magnetoresistance and strong temperature dependence of magnetization. For higher t coalescence of Fe granules occurs and metallic percolation cluster is formed at t~0.81 nm. This is manifested by rapid decrease of films resistance and formation of multipeak ferromagnetic resonance spectra. For t>1.25 nm a continuous coverage of MgO by Fe takes place. However, the morphology of Fe layers is rough. This causes the appearance of magnetostatic resonance modes analogous to those observed for continuous films deposited on embossed surfaces.

1. Introduction
Magnetic metal-insulator nanostructures and thin films were intensively studied in past decades. They show outstanding magneto-transport and magneto-optical properties which makes them promising candidates for next generation magnetic recording media, low magnetic field sensors and other spintronics applications. Granular films and multilayers of various magnetic metals (Fe, Co, Ni) and alloys (CoFe, FeNi) embedded in different insulator matrixes (SiO₂, Al₂O₃, TiO₂, ZrO₂) have been studied previously [1-8]. Multilayers, especially discontinuous metal-insulator multilayers (DMIMs), prepared by sequential deposition are of particular interest [4, 5]. DMIMs show relatively high tunnelling magnetoresistance (TMR) and enhanced low field magnetic sensitivity since two dimensional (2D) arrangements of magnetic particles favours dipolar interactions within the layer [4]. The structural state of metallic phase in DMIMs varies with increasing nominal thickness of metallic layer (t) from an ensemble of metallic granules incorporated in insulator matrix to a continuous metallic films separated by an insulator. This causes dramatic changes in physical properties. Recently
enhanced TMR values have been reported in Fe/MgO/Fe epitaxial magnetic tunnel junctions (MTJ) [9]. This effect was associated with the crucial role of band symmetry and coherent tunnelling. On the other hand, experimental data on DMIMs of Fe/MgO are generally lacking. Only a limited number of experiments on Fe/MgO/Fe trilayers and DMIMs have been carried out recently [10-13]. Isotropic TMR values of ~3.3% at room temperature and ~10% at 30K in magnetic field H=18 kOe were reported [13]. However details of Fe layer morphology and microstructure are still not clear.

Transmission electron microscopy (TEM) and X-ray diffraction are basic techniques to investigate microstructure of thin films. However, both techniques have drawbacks. X-ray reflectivity (XRR) proves that investigated Fe/MgO multilayers have periodic structure [11]. Bilayer thickness derived from the fitting of the experimental XRR profiles using LEPTOS software (BRUKER AXS) in assumption of strictly periodic structure was slightly lower than expected from the deposition rates measured for continuous films. The difference between nominal and fitted Fe/MgO bilayer thickness was about 5% for t>0.81 nm. It was supposed that the difference is caused by surface roughness. On the other hand, XRR fitting is hardly reliable in discontinuous regime (for small t values) due to non-continuous Fe/MgO interfaces. Cross-section TEM confirms that the structure of Fe layers changes from an ensemble of small granules barely distinguishable from the MgO matrix for t=0.40 nm, to continuous Fe layers for t=1.25 nm. The polycrystalline state of the MgO layers was also evident [11]. But cross-section TEM provides information only from small area of the sample. Also, the TEM specimen thickness (generally a few tens of nanometers) is much larger than typical Fe grain size (2-3 nm). As a result superimposition of the grains two dimensional projections within specimen thickness gives the impression of a continuous Fe layer. Thus, to determine the evolution of the morphology of metallic layers and establish correlations between structure and physical properties of DMIM one should use complementary experimental methods. Here we report the results of indirect structure investigations of Fe/MgO DMIMs using magnetic, transport and resonance techniques.

2. Experiment
Granular multilayers MgO(3 nm)/[Fe(t nm)/MgO(3 nm)]N (0.4 nm<t<1.50 nm, N is the number of bilayers) were deposited on glass substrates by pulsed laser deposition (PLD). The details of films preparation, structural, transport and magnetic characterizations were reported earlier [11-13]. Ferromagnetic resonance (FMR) measurements were carried out at room temperature at 9.45 GHz using a Bruker ELEXSYS-E500 spectrometer equipped with a goniometer.

3. Results and Discussion
Comparison of magnetic and transport properties is made in figure 1. The values of magnetization (M) measured at 5K and in H=50 kOe practically do not change for t>0.81. This indicates presence of continuous ferromagnetic layers in the films [11]. However M values are lower comparing to bulk Fe. The reduction was attributed to formation of mixed Fe-O layer on Fe/MgO interfaces. Such an intermixture seems to be typical for Fe based multilayers [7, 8]. Electrical resistivity of the films (ρ) shows sharp increase as t drops below 0.81 nm. This is a characteristic of metallic layer structure transition from continuous to discontinuous state [4, 10, 12]. The values of ρ for t<0.81 nm are higher comparing to bulk Fe due to effects associated with dimensionality confinements [14, 15] but correspond well to earlier published data on thin continuous Fe films [10, 16-18]. Thus, both magnetometry and transport measurements pointed to the transition from continuous to discontinuous state of Fe layers at t~0.81 nm.

Zero field cooled (ZFC) and field cooled (FC) magnetic susceptibility and magnetization measurements at different temperatures allow estimation of an average granule size (D_{avr}) for films with discontinuous Fe layers [11]. Samples with t≤0.61 nm demonstrate superparamagnetic properties. The bifurcation temperature (T_b) above which ZFC and FC curves superimpose strongly depends on t and increases from 25K to 100 K when t varies from 0.40 nm to 0.61 nm. This indicates increase of D_{avr}. For T>T_b estimations using Curie-Weiss law give D_{avr}~2 and D_{avr}~2.8 nm for t=0.4 nm and t=0.61 nm, respectively. The same results were obtained by fitting magnetization loops using weighted
sum of Langevin functions. In the latter case a broadening of the particle size distribution was also found [11]. This effect is associated with coalescence and aggregation of metallic particles in cermet systems approaching percolation threshold.

Below \( t=0.61 \) nm electron tunneling govern the conductance and isotropic TMR effect was observed. Average intergranular distance (\( s \)) for films with discontinuous Fe layers was estimated in Ref. 13 from temperature dependence of conductance using approach of Abeles et al. [1]. The values of \( s\sim 2.1 \) nm and \( s\sim 1.1 \) nm were evaluated for films with \( t=0.53 \) nm and \( t=0.61 \) nm, respectively. The derived intergranular distance is smaller than the thickness of MgO layer (3 nm). This result supports the assumption that coalescence of Fe granules and tunneling between them takes place mainly within the same layer.

For the films with \( t\geq 0.81 \) nm, \( T_s \) is above room temperature and ferromagnetic order is present in the whole temperature range. Anisotropic magnetoresistance (AMR) typical of continuous ferromagnetic films was also found [12]. For \( t\geq 1.25 \) nm predominantly metallic conductance was observed. In the range of \( 0.81 \) nm \( \leq t \leq 1.25 \) nm a crossover from the 2D disordered conductance to metallic regime takes place [12, 18]. Judging magnetic and transport properties the films within this \( t \) range could be described as 2D percolated structures for which conditions of continuous coverage are not fulfilled.

To provide additional information about the evolution of the morphology of Fe layers with \( t \) FMR technique was used. Out-of-plane angular dependences of the resonance fields as well as resonance linewidths were studied. The results are presented in figure 2. The analysis of the FMR data for such films is based on the simple Kittel formula [19]

\[
\omega^2 = \gamma^2 \left[ H + (N_x - N_y)M_s \right] \left[ H + (N_y - N_z)M_s \right]
\]

which is analogous to the one for continuous films. Here \( \omega \) is the FMR frequency, \( \gamma \) is the gyromagnetic ratio, \( M_s \) is the saturation magnetization, and \( N_i \) are the components of demagnetizing tensor in the coordinate frame with principal z-axis along the external field \( H \). However, the internal dipolar fields in nanostructured materials (especially for DMIMs) do not always fit the classical concept of demagnetizing tensor [20]. It has been shown earlier [21] that the construction of demagnetizing tensor for such systems is very similar to that for a three-dimensional granular sample:

\[
\vec{N}_{eff} = f\vec{N}_s + (1-f)\vec{N}_g
\]
where $\tilde{N}_s$ and $\tilde{N}_g$ are the tensors for sample and granule respectively, and $f$ the volume filling factor. Although the main results in [20] were obtained for square grids of granules the approach presented in that work allows qualitative analysis of our systems.

![Figure 2](https://www.example.com/figure2.png)

**Figure 2.** FMR spectra of Fe ($t$ nm)/MgO (3 nm) DMIMs. (a) $t=0.40$ nm; (b) $t=0.61$ nm; (c) $t=0.90$ nm; (d) $t=1.50$ nm. Sharp resonance line at $H\sim3500$ Oe corresponds to sample holder.

For the sample with $t=0.40$ nm the resonance line is similar to granular films with weak magnetostatic interaction between granules. There is practically no difference between the resonance fields for perpendicular and in-plane configuration (Fig. 2a). The increase of $t$ up to 0.61 nm (Fig. 2b) enlarges the splitting between perpendicular and in-plane resonance fields. This should be ascribed to the boost of magnetostatic interaction between granules. It is worth noting that the resonance linewidth increases twice comparing to the sample with $t=0.40$ nm (1700 Oe and 780 Oe for $t=0.61$ nm and $t=0.40$ nm, respectively). It is clearly seen from Eq. 2 that the influence of individual granule contribution to the resonance line broadening decreases with decreasing distances between granules (increase of the volume filling factor). This mechanism should cause reduction of the resonance linewidth with the increase of the filling factor for the granules of the same shape. At the same time the decrease of the distances between granules in disordered system leads to the increase in dispersion of magnetostatic fields on granules coming from their local environment, which is not taking into
account in [20]. Also one should consider that the increase of granules concentration in layers leads to their coalescence. As a result, the latter reasons associated with the increase of dispersion of demagnetizing factors due to the particle shape distribution favor the broadening of resonance linewidth.

The dramatic decrease of the resonance linewidth (down to 450 Oe) is observed for the film with \( t = 0.9 \text{ nm} \) (Fig. 2c). This manifests an aggregation of granules in the layers and formation of extended magnetic clusters. This result is in good agreement with magnetic and transport measurements that demonstrate formation of percolation cluster of Fe in the films.

Presence of continuous ferromagnetic layer is confirmed by FMR for \( t > 1.25 \text{ nm} \). The resonance peaks are narrow (~70 Oe in the in-plane configuration) and their linewidth are close to those of continuous thick iron films. However, FMR spectra even for the film with the thickest Fe layer \( (t = 1.50 \text{ nm}, \text{Fig. 2d}) \) are not identical to those of thick films. First of all, the effective magnetization \( (M_{\text{eff}}) \) of the film derived from the angular dependences of resonance field is more than twice smaller (~650 emu/cm\(^3\)) comparing to one of continuous iron layer (1740 emu/cm\(^3\)). The decrease of \( M_{\text{eff}} \) is usually ascribed to formation of perpendicular anisotropy \( (H_{A\perp}) \) due to the layer surfaces and magnetoelastic contribution. We can not also exclude the reduction of effective demagnetizing factor of Fe layers as a result of the surface roughness, which leads to decrease of perpendicular to the film plane demagnetizing factor \( N_{\perp} \) and increase of in-plane ones \( N_{\parallel} \). Both these contributions are symmetrically entering into the resonance equations and can not be separated from FMR experiments; only the value of \( 4\pi M_{\text{eff}} = (N_{\perp} - N_{\parallel})M_s - H_{A\perp} \) can be derived. The film roughness is confirmed by multipeak FMR spectra which are analogous to those observed for continuous films deposited on embossed surfaces [22]. However these mechanisms can hardly explain such a strong decrease of \( M_{\text{eff}} \) (see for instance [23]). On the other hand the results of magnetometry measurements show a significant reduction in saturation magnetization comparing to bulk iron. Considering the fact that intermixed Fe-O layer likely exists on the Fe/MgO interface we should suppose the formation of a surface layer with low magnetization. The exchange interaction between disordered surface and internal iron atoms leads to formation of magnetic layer with reduced saturation magnetization [24]. However it seems the main mechanisms of \( M_{\text{eff}} \) reduction are the surface anisotropy and magnetoelastic contribution. Taking into consideration plausible values of effective magnetization reduction due to surface roughness and partial oxidation of metal layers the perpendicular anisotropy field can be evaluated as 9-10 kOe.

### 4. Conclusions

The combination of magnetometry, FMR and transport measurements allows to follow the evolution of the morphology of iron layers in Fe(t nm)/MgO(3 nm) with \( t \). The increase of \( t \) from 0.40 nm to 1.50 nm leads to a modification of the films from DMIMs with practically non-interacting metallic granules separated by oxide \( (t = 0.40 \text{ nm}) \) through percolated structures \( (t = 0.81 \text{ nm}) \) to multilayers with continuous metallic layers \( (t > 1.25 \text{ nm}) \). The comparison of FMR and magnetometry data allows to reveal a formation of an oxidized shell with weak magnetization on Fe/MgO interface that cause the reduction of saturation magnetization of Fe layers. The multipeak FMR spectra of continuous films for \( t \) up to 1.50 nm give evidence that the surfaces of the Fe layers are very rough. It has been shown a formation of strong perpendicular to the films plane anisotropy due to the layers surfaces and magnetostriction. However additional investigations are needed to separate these contributions.

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References

[1] Abeles B, Sheng P, Coutts M D and Arie Y 1975 Adv. Phys. 24 401
[2] Niklasson G A and Granqvist C G 1984 J. Appl. Phys. 55 3382
[3] Fujimori H, Ohnum S, Kobayashi N and Matsumoto T 2006 J. Magn. Magn. Mater. 304 32
[4] Kakazei G N, Pogorelov Yu G, Lopes A M L, Sousa J B, Cardoso S, Freitas P P, Pereira de Azevedo M M and Snoeck E 2001 J. Appl. Phys. 90 4044
[5] Dieny B, Sankar S, McCartney M R, Smith D J, Bayle-Guillemaud P and Berkowitz A E 1998 J. Magn. Magn. Mater. 185 283
[6] Vovk A, Wang J Q, He J, Zhou W, Pogoriliy A, Shypil’ O, Kravets A and Khan H 2002 J. Appl. Phys. 91 10017
[7] Dempsey N M, Ranno L, Givord D, Gonzalo J, Serna R, Fei G T, Petford-Long A K, Doole R C and Hole D E 2001 J. Appl. Phys. 90 6268
[8] Auric P, Micha J S, Proux O, Giacomoni L and Regnard J R 2000 J. Magn. Magn. Mater. 217 175
[9] Parkin S S P, Kaiser C, Panchula A, Rice P M, Hughes B, Samant M and Yang S H 2004 Nature Materials 3 862
[10] Arita M, Wakasugi K, Ohta K, Hamada K, Takahashi Y and Choi J B 2008 Microelectronic Engineering 85 2445
[11] García-García A, Vovk A, Pardo J A, Štrichovaneck P, Magén C, Snoeck E, Algarabel P A, De Teresa J M, Morellón L and Ibarra M R 2009 J. Appl. Phys. 105 063909
[12] García-García A, Vovk A, Štrichovaneck P, Pardo J A, Magén C, Algarabel P A, De Teresa J M, Morellón L and Ibarra M R 2010 J. Phys.: Condens. Matter. 22 056003
[13] García-García A, Vovk A, Pardo J A, Štrichovaneck P, Algarabel P A, Magén C, De Teresa J M, Morellón L and Ibarra M R 2010 J. Appl. Phys. 107 033704
[14] Fuchs K 1938 Proc. Camb. Phil. Soc. 34 100
[15] Sondheimer E H 1952 Adv. Phys. 1 1
[16] Schad R, Belien P, Verbanck G, Moschchalkov V V and Bruynseraede Y 1998 J. Phys.: Condens. Matter 10, 6643
[17] Sangiao S, Morellon L, Simon G, De Teresa J M, Pardo J A, Arbiol J and Ibarra M R 2009 Phys. Rev. B 79 014431
[18] Liu C, Park Y and Bader S D 1991 J. Magn. Magn. Mater. 111 L225
[19] Kittel C 1996 Introduction to Solid State Physics (Wiley, New York)
[20] Kakazei G N, Pogorelov Yu G, Costa M D, Golub V O, Sousa J B, Freitas P P, Cardoso S and Wigen P E 2005 J. Appl. Phys. 97 10A723
[21] Dubowik J 1996 Phys. Rev. B 54 1088
[22] Vovk A, Malkinski L, Golub V, Whittenburg S, O’Connor C, Jung J S and Min S H 2005 J. Appl. Phys. 97 10J506
[23] Schlömann E 1970 J. Appl. Phys. 41 1617
[24] Guslienko K Y, Lesnik N A, Mitsek A I and Vozniuk B P 1991 J. Appl. Phys. 69 5316