On the exchange coupling interaction between Fe and L1₀-ordered FePd thin films

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Abstract. Hard L1₀-ordered FePd ultra-thin films were epitaxially grown by means of a molecular beam epitaxy system on MgO-(100) monocrystalline substrates. Because of the very low thickness of the films, small amounts of superparamagnetic L1₀-FePd particles were also formed on the surface. The magnetic properties of the films are typical of highly coercive systems showing a strong perpendicular anisotropy. These hard films were used for the development of soft/hard exchange-spring magnets, by depositing different thickness of iron. Fe reacts with the surface FePd small particles giving rise to a Fe(Pd) compound with a broad distribution of particle sizes. The resulting systems are bi-layers constituted by a soft Fe(Pd) and a hard L1₀-FePd layer intercalated by a very thin region of small particles. For soft/hard thickness ratio nominally equal to 1 the magnetic behaviour is typical of a single-phase hard magnet. By increasing the nominal thickness ratio up to 3, the behaviour becomes typical of an exchange-coupled magnet with hysteresis loops characterized by two critical fields.

1. Introduction

FePd thin films grown at room temperature show a fcc structure (A1 phase) characterized by soft magnetic properties. By growing the films at high temperatures or by means of post-annealing treatments the alloy around the equiatomic composition undergoes a structural transformation with the establishment of a fct structure (L1₀ phase), characterized by a strong uniaxial magnetocrystalline anisotropy parallel to the contracted c-axis. Therefore, by means of an epitaxial growth it is possible to obtain systems showing perpendicular anisotropy [1-3]. Contrary to L1₀-ordered FePt and CoPt, the FePd is reported to show a much lower coercivity and squareness [4-8]. For this reason, the L1₀-ordered FePd alloy has been evaluated not to be suitable for the development of exchange-spring magnets, i.e. systems where soft and hard magnetic phases are intercalated and exchange-coupled at the interface, showing a single phase magnetic behaviour.

In the present work FePd thin films grown by means of an UHV e-beam evaporation technique have been obtained, showing a strong perpendicular anisotropy, high coercivity and loop squareness. These peculiar characteristics allowed to use these films as the hard phase for the development of FePd/Fe exchange-spring magnets having a high value of the soft/hard thickness ratio. The obtained results are quite unexpected and surprising considering that (i) no data are reported about FePd systems showing values of the coercivity up to 10 kOe and (ii) in literature, the coupling among the different magnetic phases is reported to require a soft/hard thickness ratio lower than 1.
2. Experimental

FePd thin films, 4.5 nm thick, were grown in ultra-high vacuum by means of a molecular beam epitaxy (MBE) system equipped with electron gun evaporators. The FePd films were deposited onto MgO-(100) monocrystalline substrates at 540°C, using iron highly enriched in the $^{57}$Fe Mössbauer isotope. On the top of the FePd films, $^{57}$Fe layers of different thickness were deposited at room temperature. In the following these samples were labelled as Fe($t_{Fe}$/FePd, where $t_{Fe}$ = 4.5, 10 and 15 is the Fe thickness in nm.

Conversion electron Mössbauer spectroscopy (CEMS) measurements were performed without breaking vacuum, using a 50 mCi $^{57}$Co(Rh) source with the $\gamma$-ray incident angle of 17°. The isomer shifts are referred to $\alpha$-Fe, and the fitting was carried out using a least-square minimization routine with a combination of linear and non-linear regressions as a superposition of Lorentzian lines. For the fittings, the intensity ratio between the 1st (6th) and the 3rd (4th) lines has been maintained equal to 3. The magnetic properties were measured with a magneto-optical Kerr effect magnetometer (MOKE) using a $s$-polarized 633 nm laser light with the magnetic field applied both perpendicular (polar geometry) and along the film plane (longitudinal geometry). The dcd remanence curves were measured in polar geometry starting with the sample in the saturated state. Ultra-high vacuum atomic and magnetic force microscopy (AFM, MFM) was used in tapping/lift mode to characterize the morphology and domain patterns of the samples. For MFM, the tip-sample distance was 50 nm.

3. Results and Discussion

Figure 1(a) shows the Mössbauer spectrum measured for the FePd films grown at 540°C. It can be fitted as the superposition of a quadrupole doublet ($\delta$) and a series of sextets ($\nu$) whose hyperfine field distribution $H_{hf}$ is reported in the inset. The ferromagnetic contribution, showing a mean $H_{hf}$ = 268 kOe and a quadrupole splitting $\Delta E_Q$ = 0.20 mm/s, is due to the L1$_0$-ordered FePd alloy having an almost equiatomic composition [2]. Taking into account the $\gamma$-ray incident angle, the line intensity ratios (3:1.3:1) indicate that the mean magnetization vector is oriented at ~25° from the perpendicular to the film plane. As for the quadrupole doublet ($\Delta E_Q$ = 1.08 mm/s), it can be attributed to the same L1$_0$-FePd in the form of small particles showing a superparamagnetic behaviour [9]. Accordingly, the AFM measurements show a morphology constituted by irregular and disconnected islands as well as small droplets having nanometric sizes.

The corresponding MOKE measurements performed in polar geometry are reported in figure 1(b). The film shows a well square-shaped hysteresis loop with a remanence to saturation ratio $M_r/M_s$ of 92% and a coercive field $H_c = 2.2$ kOe. The presence of a strong perpendicular anisotropy and the absence of appreciable in-plane contributions to the magnetization vector are confirmed by the almost zero-remanence of the longitudinal loop (figure 2).

No magnetic morphology was evidenced by the MFM analysis. This fact may well be due to the presence onto the sample surface of the superparamagnetic small particles. Moreover, the low thickness of the films as well as the strong perpendicular anisotropy of the ferromagnetic material can give rise to very large domain sizes, well overcoming the maximum scanning area of the adopted microscope (~5 x 5 $\mu$m).

Figure 1(c) shows the CEMS spectrum measured after covering the film with a 4.5 nm thick iron layer, Fe(4.5)/FePd. The spectrum was fitted superimposing, to the $\nu$ and $\delta$ contributions due to the hard FePd film, a series of sextets ($\chi$) whose $H_{hf}$ distribution is reported in the inset. The fitting procedure was performed maintaining the values of the $\nu$ and $\delta$ hyperfine parameters equal to those found for the FePd spectrum [figure 1(a)], and leaving free to change only the relative areas of the two components.

Considering that no appreciable contribution from pure iron is present in the spectrum and that the $\delta$ contribution due to FePd small particles is considerably reduced, it is possible to assume that Fe interacted with the highly reactive FePd small particles giving rise to Fe containing low amounts of Pd, Fe(Pd), showing a broad distribution of particle sizes ($\chi$ contribution). This hypothesis means that, as a consequence of the Fe deposition, the sample grew in the form of a bi-layer constituted by a
superficial soft Fe(Pd) layer and a hard L1_0-FePd layer, intercalated by a very thin interfacial region of superparamagnetic particles. The effect of the Fe(Pd) growth in reducing the amount of FePd small particles with the formation of a continuous surface layer is supported by the AFM image of the bi-layer [figure 3(a)].

![Figure 1](image)

**Figure 1.** CEMS spectra (left column) and polar dcd recoil loops (right column) measured for (a) and (b) FePd, (c) and (d) Fe(4.5)/FePd and (e) and (f) Fe(15)/FePd samples. The hyperfine magnetic field distributions corresponding to the contributions to the CEMS spectra and the initial curves are reported in the insets.

From the line intensity ratios of the χ contribution it follows that the angle between the magnetization vector and the perpendicular to the film plane is ~22°. This low tilting is attributable to a strong coupling between the soft Fe(Pd) and the hard L1_0-FePd layer. In agreement, the bi-layer hysteresis loop [figure 1(d)] is typical of a single-phase hard magnet characterized by a high squareness due to a strong perpendicular anisotropy. The decrease of the FePd superparamagnetic particles determines an appreciable increase of the coercivity up to 2.6 kOe. The first magnetization curve suggests that the coercivity is mainly controlled by the nucleation of reversed domains which gives rise to the abrupt rotation of the magnetic moments for very low applied fields.

The low anisotropy of the superficial Fe(Pd) compound and the decrease of the FePd small particles present at the Fe(Pd)/L1_0-FePd interface give rise to a magnetic morphology constituted by irregular small domains [figure 3(b)]. The high image contrast and the irregularity of the domain pattern can only be attributed to the strong perpendicular anisotropy characterizing the bi-layer.

For the Fe(15)/FePd, the Mössbauer spectrum [figure 1(e)] shows that the amount of Fe(Pd) is appreciably increased (set of sextets χ), while no contribution from pure iron is detectable. The presence of the δ doublet shielded by the χ contribution was also considered, but its introduction...
among the fitting components gave no improvements, tending on the contrary to increase the standard deviation. These facts reasonably mean that all deposited iron and FePd small particles reacted giving rise to the Fe(Pd) compound.

The angle between the Fe(Pd) magnetization vector and the perpendicular to the film plane increases to 50°. Correspondingly, a significant decrease occurs of both the film coercivity (1.95 kOe) and the squareness (32%) of the hysteresis loop [figure 1(f)]. Two critical fields for the magnetization reversal establish, i.e., the nucleation, $H_N$, and the demagnetizing field, $H_D$. For applied fields in the $H_N \div H_D$ range, the rotation of the magnetic moments is completely reversible, indicating that a good exchange coupling established between the hard L1$_0$-FePd and the soft Fe(Pd) layer. The difficulty to reach the complete saturation can be ascribed to the in-plane component of the magnetization vector.

In effect, while in the L1$_0$-FePd hard layer the magnetization vector is oriented perpendicular to the film plane, the deposition of Fe determines the appearance of an appreciable contribution oriented into the film plane whose strength increases by increasing the Fe deposition thickness (figure 2). However, also for the Fe(15)/FePd the strength of this in-plane contribution, as well as of the demagnetizing field, are not sufficient to completely overcome the L1$_0$-FePd perpendicular anisotropy. In agreement, the MFM measurements [figure 3(d)] show a well defined magnetic morphology constituted by small domains. The relatively low image contrast may well be connected to the in-plane component of the magnetization vector.

![Figure 2](image2.png)

**Figure 2.** Longitudinal hysteresis loops for the FePd hard film, Fe(4.5)/FePd and Fe(15)/FePd.

![Figure 3](image3.png)

**Figure 3.** AFM and MFM images (4 × 4 µm) for Fe(4.5)/FePd (a, b) and Fe(15)/FePd (c, d).
4. Conclusions

FePd thin films 4.5 nm thick were grown in ultra-high vacuum onto MgO monocrystalline substrates at 540°C by means of a MBE equipped with electron guns. The adopted conditions allowed to obtain an epitaxial growth of a well ordered L1₀ phase with the c-axis perpendicular to the film plane. The films show a strong perpendicular anisotropy with hysteresis loops characterized by a very high squareness and coercivity. Surface Mössbauer measurements evidenced that the L1₀-FePd is present not only in its ferromagnetic form with a strong perpendicular texture, but also in the form of small particles showing a superparamagnetic behaviour.

When iron is evaporated onto these hard films, a reaction occurs with the highly reactive superparamagnetic FePd particles, giving rise to a Fe-base compound containing small amounts of Pd, Fe(Pd). Therefore, the resulting systems are bi-layers constituted by the soft Fe(Pd) and the hard L1₀-FePd layer intercalated by a very thin region of small particles.

For a Fe deposition up to 4.5 nm, the bi-layers show a magnetic behaviour typical of a single-phase hard magnet with hysteresis loops characterized by a high squareness and a coercivity slightly higher than that of the original hard phase. By increasing the Fe deposition, the Fe(Pd) thickness progressively increases, and, even for a soft/hard ratio nominally equal to 3, a good exchange coupling establishes between the different phases giving rise to a single-phase magnetic behaviour. However, the high value of the soft to hard thickness ratio determines both a decrease of coercivity and the appearance of two critical fields for the magnetization reversal.

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