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Compositional dependence of the magnetic properties of epitaxial FeV/MgO thin films

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Owing to their bcc structures and the low lattice misfit with MgO, FeV alloys are interesting for MgO-based magnetic tunnel junctions. We use vector network analyzer ferromagnetic resonance to measure the magnetization, anisotropy, exchange stiffness, and damping of epitaxial FeV/MgO thin alloys of various V contents. The low magnetization, very high exchange stiffness (23 pJ/m) and very low effective damping (<0.0026) of the alloy with 20% V content makes it an interesting candidate for spin torque applications. The ultralow damping is consistent with a spin-orbit origin, which sheds light on the possible strategies to reduce the damping in alloys. © 2013 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4845375]

Magnetic tunnel junctions form the base element of many spintronic devices. One of the most emblematic application is the spin torque operated magnetic random access memory,1 that is presently undergoing a shift from in-plane magnetized systems2 to out-of-plane magnetized systems.3 In this emerging “perpendicular” technology, the storage layer needs to warrant a high tunnel magneto resistance (TMR) ratio, a sufficient perpendicular anisotropy, and a lower switching current. This requires to be able to crystallize the storage layer into a body-centered cubic (bcc) structure with a (001) texture, and a reasonably good lattice matching with MgO. In addition, the storage layer material also needs to have a large exchange anisotropy with MgO and a low Gilbert damping. As a result, CoFeB alloys are usually chosen,3-5 because they have the relevant properties.6,7 However intermetallics with greater exchange stiffness and Curie temperature, together with lower magnetization $M_s$ and lower damping are desirable. This is first to fully benefit from the interface anisotropy without paying the destabilization cost of the perpendicular demagnetization field and second, to favor coherent magnetization reversal compared to domain wall mediated reversal.8

Most of the research activity has been focused on the alloying of Fe, with the aim of starting from a bcc structure and a damping much lower than Co and Ni.9 Prior trials to add Cr, V, or Ni dopants in FeCo or FeNi systems10,11 were found to degrade substantially either the damping factors or the TMR ratios. One thus still needs to find a material, or a material combination yielding together large TMR, low damping and low magnetization.

To find one, our reasoning is the following. Reducing the magnetization can be done by substituting iron by other transition metal elements, following the Slater-Pauling rule. To avoid miscibility problems, we should preferentially substitute with materials ordering in a bcc state (V, Cr, Mn, Nb, Mo, Ta, W) when being elemental solids, or making solid solutions with bcc Fe on the Fe-rich side. This excludes the noble metals (Au, Cu, Ag) and Ni. Our second guiding line is the search for lower Gilbert damping parameters $\alpha$. Unfortunately, $\alpha$ is one of the last few features that cannot be calculated reliably in transition metal alloys. However, $\alpha$ is known12,13 to vary like the square of the spin-orbit constant $\zeta$ provided that the band structure and the Fermi level are unchanged.14 Within a transition series, the spin-orbit constant varies15 like $Z^2$, and from one transition series to the next it increases substantially.16 A direct consequence is that one should avoid substituting iron with elements from the 4d and 5d series, and from the end of the 3d series. We are thus left essentially with V, Cr, and Mn. Mn can be excluded since it gets oxidized when in proximity with MgO,17 which leads to poor magnetic properties. Being the lightest element, Vanadium is the best candidate for an effective reduction of the magnetization, together with a decrease of the damping.18

Another uncommon18,19 fact that is often forgotten is that the Curie temperature of FeV alloys increases with the Vanadium doping.20,21 The Curie temperature is maximum for a Vanadium content of 20%. At that composition, the exchange integral between Fe atoms was predicted to be twice the value of pure iron,20 which should favor a more coherent magnetization reversal. Another advantage of FeV alloys compared to CoFe is their small misfit with the MgO lattice. This was shown to reduce the density of misfit dislocations in the MgO barrier,22 with a significantly increase of the TMR ratio.23 Finally, perpendicular magnetic anisotropy24 with some electric field tunability25 was obtained formerly in Fe/V systems which led us to the conclusion that the potential FeV alloys deserve to be assessed.

We have prepared MgO (single-crystalline substrate)/Fe$_{1-x}$V$_x$ (9–30 nm)/Au (6 nm) films, with 60% $\geq x \geq 0%$ by molecular beam epitaxy. Separate V and Fe layers of adequate thicknesses are first grown at room temperature in epitaxy with the MgO, as checked by RHEED patterns.22 This is followed by an 800 °C annealing, which yields an efficient mixing of Fe and V, attested by the symmetry of the (002) X-ray diffraction peak (not shown). The Fe$_{1-x}$V$_x$ layers have bcc

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structure with their [100] directions rotated by 45° with respect to the MgO cubic axes. The V contents x were checked by the modeling of X-ray photoelectron spectra at the 2p lines of Fe and V, as detailed in Ref. 23. The contents were found to be consistent with the lattice spacing of the alloy as deduced from X-ray diffraction. The film thicknesses t were checked by the modeling of Grazing X-ray Reflectometry spectra. An addition series of samples has been used to deduce anisotropy field. In principle, these two quantities can be measured in zero field when the magnetization lies along the RF field and compare with the VNAFMR outcomes. For the Fe50V50 and Fe58V42 compositions only a broadening contribution. Inset: PMOKE loop showing the recovery of a physical susceptibility, which was obtained with vanishing susceptibility, and is the exchange stiffness and \( H_{\text{sat}} = S - H_{Kc} \) where \( S \) is the magnetization and \( H_{Kc} \) the anisotropy field.

To quantify the magnetic properties, we have used VNAFMR to obtain the sample’s high-frequency (1 to 70 GHz) transverse susceptibility \( \chi_t \) (Fig. 1). We use the open-circuit total reflection configuration27 with an applied field \( \mu_0 H_t \) of 0–2.5 T perpendicular to the sample surface. To deduce the complex permeability \( \mu_{\text{eff}} \) of the effective medium surrounding the strip line, we need a reference spectrum with vanishing susceptibility, which was obtained in zero field when the magnetization lies along the RF field radiated by the strip line. While for ultrathin films the sample transverse susceptibility \( \chi_t \) is strictly proportional to the \( \mu_{\text{eff}} \), this is not the case for thick metallic films because eddy current screening in the film mixes the real and imaginary parts of the sample permeability in a frequency dependent manner.28 For small FMR linewidths, the recovery of a physical susceptibility can be done by projecting \( \Re(\mu_{\text{eff}}) \) and \( \Im(\mu_{\text{eff}}) \) on symmetric and antisymmetric Lorentzian functions as exemplified in Figure 1.

To analyze the characteristic features in the susceptibility spectra, we consider films with cubic anisotropy and easy axis in the film plane. The in-phase susceptibility \( \Re(\chi_t) \) vanishes at the ferromagnetic resonance frequency \( \omega_{\text{FMR}} / 2\pi \). For fields \( |H_z| \geq H_{\text{sat}} \), i.e., saturating the magnetization along \( z \), we have \( \omega_{\text{FMR}} = \gamma_0 (|H_z| - M_S + H_{Kc}) \), where \( \gamma_0 \) is the gyromagnetic ratio. \( \chi_t \) also vanishes at the frequencies of the perpendicular standing spin waves (PSSW) when the phase difference between dynamic magnetizations at the top and bottom surfaces is \( \pi \). For fields \( H_z \geq H_{\text{sat}} \) and in the absence of surface pinning, this happens at frequencies

\[
\omega_{\text{PSSW}} = \omega_{\text{FMR}} + \frac{\gamma_0}{\mu_0 M_s} \frac{2 \Delta n^2}{A},
\]

where \( A \) is the exchange stiffness and \( n \) the mode index which is an integer in the case of pinning-free surfaces. Unfortunately the \( n \geq 3 \) PSSW modes are probably too high in frequency to be observed with our technique (Fig. 2(c)). Linear fits of either \( \omega_{\text{FMR}} \) or \( \omega_{\text{PSSW}} \) versus \( H_z \) for fields above \( H_{\text{sat}} \) are used to get \( \gamma_0 \), which is then translated in a spectroscopic splitting factor \( g \). The zero-field intercept of the curve is then used to deduce \( M_S - H_{Kc} \). The film resonance frequency at remanence is \( \gamma_0 \sqrt{H_z (M_S + H_{Kc})} \). Its knowledge, combined

\[ \text{FIG. 1. Permeability spectrum recorded on a Fe(20 nm)/MgO film at an applied field of 2.1 T (red dots) perpendicular to the sample. The frequency interval is chosen to display the first (n = 1) perpendicular standing spin wave mode. The fit (bold black line) is done with an effective linewidth parameter } \Delta \omega / (2 \omega) = 0.0047, \text{ which includes an unknown inhomogeneous broadening contribution. Inset: PMOKE loop showing } M_t, \text{ versus } \mu_0 H_t, \text{ for the same film.} \]

\[ \text{FIG. 2. (A) Magnetization versus Fe content. The dotted line is the Slater-Pauling rule. (B) Anisotropy versus Fe content. The line is a guide to the eye. (C) Field dependence of the Ferromagnetic Resonance frequencies and the perpendicular standing spin wave frequencies of Fe/MgO films of various thicknesses.} \]
with $H_{\text{sat}}$ yields the magnetization $M_S$ and the anisotropy field $H_{\text{ka}}$. Finally, $\omega_{\text{PSSW}} - \omega_{\text{FMR}}$ is used to deduce the exchange stiffness $A$. When several PSSW modes were detectable, the $n^2$ dependence of Eq. (1) was not observed, indicating some surface pinning of the magnetization, probably related to surface anisotropy. Because the influence of surface pinning on the spin wave wavelength increases with $n$, we have chosen to use only the $n = 1$ mode to deduce the exchange stiffness $A$. The validity of this approach is yet to be clarified.

To extract the damping, we only use the high field, i.e., $|H_z| \geq H_{\text{sat}}$ data. Indeed, when the magnetization is saturated along $z$, two-magnon scattering is forbidden, such that the FMR linewidth is only influenced by Gilbert damping $x$ and long-range inhomogeneities of the internal field (so-called inhomogeneous broadening). The positive peak to negative peak frequency spacing of $\Re[\mu_e]$, or equivalently the full width at half maximum of the peak of $\Im[\mu_e]$ is $\Delta \omega = 2\omega_{\text{FMR}}$. Each line can thus provide an upper bound for $x$, and the frequency dependence of the linewidth (Fig. 3(a)) is an indication of the inhomogeneity of the internal field in the material. Note that for the Fe$_{50}$V$_{50}$ composition an extreme broadening was observed, as two clearly distinct FMR lines were seen (Fig. 3(b)). We report here only the results for the most intense line. The weakest mode corresponds to a region in the sample where the magnetization is reduced by 13 mT.

The magnetic properties of our films are gathered in Table I. The Landé factors are scattered in the interval $2.06 \leq g \leq 2.10$ which is essentially our error bar; they thus stay near that of pure iron ($g = 2.09$). The magnetization (Fig. 2(a)) is in line with expectation, decreasing with the Vanadium content. The magnetization falls on the Slater-Pauling curve $\mu_0 M_S = 2.2 - 3x$ till $x \approx 40\%$. For larger V content, the magnetization falls off faster, and the sample is paramagnetic at room temperature for $x = 60\%$. The magnetizations for $x \geq 40\%$ are thus influenced by the proximity of Curie temperature $T_C$ and room temperature. The cubic anisotropy field (Fig. 2(b)) also decreases with the Vanadium content, at a rate faster than the magnetization. It changes sign for about 40% of Vanadium and more, and we shall see that the sample properties get less uniform for these compositions.

More surprising is the dependence of the exchange stiffnesses of the alloys (Table I). Indeed, the exchange stiffness is essentially independent of the Vanadium content as long as $T_C \gg 300$K, i.e., for $x \leq 40\%$. This is opposite to common thinking that would bet on a reduction of the exchange stiffness with the dilution of Fe by the nonmagnetic V. This rather constant $A$ has important consequences: a qualitative estimate of the average exchange integral in a material of given fixed structure (bcc here) is provided by the exchange length. In Table I, we can observe a very clear increase of the exchange length with the Vanadium content. To some extent, this is consistent with the increase of Curie temperature observed in FeV alloys for V contents up to $x \approx 20\%$. This increase in exchange length is technically interesting in storage applications, as it hinders domain wall based reversal that reduce the thermal stability of the magnetization.

The last important parameter is the damping parameter. In most of our samples, the Gilbert damping is so low that the FMR resonance lines include inevitably some inhomogeneous broadening that is difficult to separate from the Gilbert damping contribution. Also for such low damping values, the contributions of Eddy currents are known not to be negligible in our range of thicknesses especially for single-crystalline films like ours. We thus report (Table I) only upper bounds of the Gilbert damping, by saturating the magnetization in the perpendicular direction and using $\Delta \omega_{\text{max}} = \Delta \omega / (2 \omega_{\text{FMR}})$. Star symbols indicate when the frequency dependence of $\Delta \omega / (2 \omega_{\text{FMR}})$ is indicative of substantial inhomogeneous

<table>
<thead>
<tr>
<th>Composition</th>
<th>$\mu_0 M_S$ (T)±0.03</th>
<th>$H_{\text{KA}}$ (mT)±2</th>
<th>$\Delta \omega_{\text{max}}$ ±10%</th>
<th>$\sqrt{\frac{\chi}{\mu_0}}$ (nm)</th>
<th>$A$ (pJ/m)±2</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe/MgO</td>
<td>2.2</td>
<td>56</td>
<td>≤ 0.0047</td>
<td>3.5</td>
<td>23</td>
</tr>
<tr>
<td>Fe$<em>{50}$V$</em>{10}$</td>
<td>1.93</td>
<td>52</td>
<td>≤ 0.0055</td>
<td>4</td>
<td>22</td>
</tr>
<tr>
<td>Fe$<em>{50}$V$</em>{15}$</td>
<td>1.72</td>
<td>45</td>
<td>≤ 0.0070</td>
<td>4.6</td>
<td>26</td>
</tr>
<tr>
<td>Fe$<em>{50}$V$</em>{20}$</td>
<td>1.62</td>
<td>40</td>
<td>≤ 0.0026</td>
<td>4.7</td>
<td>23</td>
</tr>
<tr>
<td>Fe$<em>{75}$V$</em>{25}$</td>
<td>1.45</td>
<td>32</td>
<td>≤ 0.0060</td>
<td>5.0</td>
<td>22</td>
</tr>
<tr>
<td>Fe$<em>{84}$V$</em>{16}$</td>
<td>1.25</td>
<td>30</td>
<td>≤ 0.011(*)</td>
<td>6.7</td>
<td>26</td>
</tr>
<tr>
<td>Fe$<em>{94}$V$</em>{6}$</td>
<td>1.2</td>
<td>19</td>
<td>≤ 0.0080</td>
<td>5.9</td>
<td>24</td>
</tr>
<tr>
<td>Fe$<em>{67}$V$</em>{33}$</td>
<td>0.7</td>
<td>-9</td>
<td>≤ 0.028(*)</td>
<td>7.2</td>
<td>10</td>
</tr>
<tr>
<td>Fe$<em>{58}$V$</em>{42}$</td>
<td>0.38</td>
<td>-5</td>
<td>≤ 0.01(*)</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Fe$<em>{50}$V$</em>{50}$</td>
<td>0</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Bulk Fe</td>
<td>2.2</td>
<td>55</td>
<td>0.0019 (Ref. 13)</td>
<td>3.2</td>
<td>20.0 (Ref. 35)</td>
</tr>
</tbody>
</table>

FIG. 3. (a) Half linewidth versus resonance frequency for a Fe$_{50}$V$_{20}$ film with 18 nm thickness. The line is a linear fit of slope 0.002 and zero frequency intercept 43 MHz. (b) Imaginary part of the permeabilities of FeMgO (20 nm)/Au (6 nm) films, and comparison with iron literature values and values of films MgO (substrate)/FeV (30 nm)/Ru (3 nm)/Pt (4 nm) deposited by sputtering († symbol). In the damping column, the stars (*) indicate the cases where a substantial inhomogeneous broadening prevents the estimation of the damping. The - symbols recall the samples for which that the PSSW mode could not be detected.
broadening. While the damping values are tarnished with some uncertainty, they prove that the damping of FeV alloys can be substantially below that of CoFeB alloys\(^6\) which never fall below 0.004 even for large thicknesses.\(26,31,32\) We illustrate these exceptionally narrow linewidths in Figure 3(b), where we have gathered the imaginary parts of the permeabilities of our 20 nm thick layers for saturating the PPF SPINEL program of the "Université Paris-Sud."


