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Temperature dependence of the interlayer exchange coupling in epitaxial Fe1/MgO/Fe2/Co tunnel junctions

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The temperature dependence of the interlayer exchange coupling has been investigated in epitaxial tunnel junctions Fe1/MgO/Fe2/Co/V with thin MgO layers using *X*-band ferromagnetic resonance (FMR) in the range 2–300 K. Variations of FMR parameters allow concluding that the coupling strength increases with temperature. This is in agreement with predictions of the theories considering pure tunneling mechanisms and contradicts the model of a resonant assisted tunneling related to defects in the insulator. The temperature dependence of the FMR linewidth shows the line narrowing under the sample heating. This may be due to the additional mechanism associated with the coupling. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2784942]

Advanced magnetic tunnel junctions (MTJs), consisting of Fe and Co electrodes separated by the MgO spacer, are developed for spintronics applications as magnetic sensors or random access memory elements.[1–3] From a physical standpoint, such systems are remarkable for the interlayer exchange coupling (IEC) which modifies significantly their magnetic properties.[4,5] The mechanisms of the magnetic coupling are still a subject of discussions though. A theory describing the conductance of MTJ systems has been developed by Slonczewski,[6] who proposed a model for the interlayer coupling through a tunneling barrier at $T=0$. According to recent studies, the tunnel transmission probability is strongly influenced by resonant effects either at the interfaces[7–10] or within the barrier.[11] All the theories predict an exponential decay of the IEC with a barrier thickness, but different temperature dependencies of the coupling. The IEC is expected to increase with the temperature in the framework of Bruno's free electron model,[12] as the tunneling barrier is lower at higher T. An increase of the coupling strength with the temperature is also expected from realistic electronic structure calculations, where the interfacial resonant state of Fe lies slightly above the Fermi level and could be activated by increasing temperature.[7–10] On the contrary, after Zhuravlev *et al.*,[11] a decrease of the IEC with the temperature is theoretically expected for resonant assisted tunneling due to defects (e.g., oxygen vacancies) within the MgO barrier. Thus, reliable data on the temperature dependence of the IEC would clarify the coupling nature and the tunneling mechanism. A quantitative estimation made in the framework of the free electron model[12] shows a weak change of the coupling in the range from 0 to 300 K.[5] Since magnetometry does not allow distinguishing such IEC variations on the background of temperature changes of other film parameters, in particular, the anisotropy, we use the *X*-band ferromagnetic resonance (FMR) to study the coupling in Fe/MgO/Co and Fe1/MgO/Fe2/Co MTJ systems. In the latter, the Co film has been used as hard magnetic layer to pin the top Fe layer in fully epitaxial MTJs.

These stacks were deposited using the molecular-beam epitaxy technique (see Ref. 4). Iron films were prepared by thermal evaporation from a standard Knudsen cell. Cobalt films, vanadium capping layers, MgO sublayers, and spacer layers were fabricated by means of an electron gun deposition. The thicknesses of ferromagnetic layers (d^{Fe} and d^{Co}) were measured using a stepmeter with accuracy of ±5–7%. The MgO thickness (d^{MgO}) values have been determined using the reflection high energy electron diffraction technique with an absolute uncertainty less than ±0.05 nm. As it has been proven using transmission electron microscopy, electrical, and magnetoresistance measurements, the spacer layers were prepared without pinholes and had flat interfaces.[4] The cobalt layer lattice was hexagonal close packed with the *c* axes lying in the film plane. There were two crystallographic domains rotated one from another by 90°. The epitaxial relationship is Co(11–20)[0001]‖Fe(100)[110] and Co(11–20)[0001]‖Fe(100)[1–10].

Reference samples were fabricated on MgO (100) substrates with MgO sublayers in such a way that they simulate each layer in a stack for FMR measurements. The growth conditions and the characterization of the films are described in more detail in our previous works.[4,5] Magnetization was obtained with a superconducting quantum interference device (SQUID) and alternating gradient field magnetometers.

FMR experiments have been performed using an *X*-band Varian spectrometer operating at the frequency $f \approx 9.25$ GHz in the field range of −100–2500 mT and temperature range of 2–300 K. The power of the microwave

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field ***h***, directed in most cases parallel to the sample surface, was 1 mW.

An analysis of FMR data is performed solving a well known resonance equation[13] together with equations of equilibrium. One obtains the equilibrium angles of the film magnetization M by minimizing the free energy density F. In the case of films with coupled layers $F=d_1F_1+d_2F_2+E_{\mathrm{ex}}$, where F_i and t_i are the free energy density and the thickness of the layer 1 or 2, respectively, and E_{ex} is the exchange coupling between the magnetic layers 1 and 2. F_i includes the Zeeman contribution, as well as the shape, magnetocrystalline, and uniaxial anisotropy energies. In the theory of FMR in films with coupled layers,[14] the exchange coupling energy is given by $E_{\mathrm{ex}}=-J_{12}(\mathbf{M}_1\mathbf{M}_2/M_1M_2)$, where J_{12} is the coupling parameter and $\mathbf{M}_1$ and $\mathbf{M}_2$ are the magnetizations of the layers 1 and 2. The theory[14] gives a prediction on the behavior of acoustic (H_r^{acoust}) and optical (H_r^{optic}) resonance modes occurring under the IEC effect instead of separate iron (H_r^{Fe}) and cobalt (H_r^{Co}) [or iron/cobalt ($H_r^{\mathrm{Fe/Co}}$)] resonances. The mode positions and intensities depend on a sign and strength of the coupling. In other words, a difference between acoustic and optical resonance fields $\delta=H_r^{\mathrm{acoust}}-H_r^{\mathrm{optic}}$ and the signal intensity ratio $I^{\mathrm{acoust}}/I^{\mathrm{opt}}$ are a measure of the IEC strength. Both of them are expected to be practically insensitive to temperature variations of the magnetic parameters of MTJ layers as, according to Ref. 14, the fields and intensities should depend rather on differences of layer magnetization and anisotropy field values, than on M_i and H_{Ai} directly. However, quantitative assessments of J_{12} may be incorrect as the model[14] does not take into account neither the influence of the ferromagnetic layer thickness[15] nor the effect of the ferromagnetic material and its electronic state[16] on the IEC.

According to magnetometric data, both soft and hard layers of stacks present fourfold symmetry, with the same directions for the easy axes. Similar to iron films,[15] the parallel resonance spectra of cobalt epitaxial samples at 9.25 GHz consist of two lines, if the film is magnetized along the *hard* magnetic axis. The lines correspond to unsaturation and saturation regimes, associated with the large magnetocrystalline anisotropy field $\boldsymbol{H}_A$. Meanwhile, along the easy axes and other directions, there is no resonance signal in positive applied fields. The bilayer $\mathrm{Fe}_5/\mathrm{Co}_{35}/\mathrm{V}_{10}$, which represents a second MTJs electrode, demonstrates a strong magnetic coupling between iron and cobalt layers and hence, weighted mean magnetic parameters, as has been estimated by FMR. It has been found by SQUID that Fe and Co film magnetizations $M_s^{\mathrm{Fe}}\cong 1700$ G and $M_s^{\mathrm{Co}}\cong 1400$ G and remain almost unchanged in the temperature range from 300 to 2 K. At 2 K, the resonance fields of iron, cobalt, and iron/cobalt electrodes increase by 10%, 60%, and 50%, respectively. Yet the temperature increase of H_r^{acoust} in MTJs is about 10% and H_r^{opt} decreases by 6% (see Fig. 2).

A unidirectional shift of the minor hysteresis loops, obtained using SQUID in both stacks Fe/MgO/Co/V and Fe1/MgO/Fe2/Co/V, indicates an antiferromagnetic (AF) coupling in agreement with FMR data shown below. The coupling parameter J_{12}, calculated using the values of exchange fields determined from the hysteresis loops, is in the range of -0.26–$0.01\ \mathrm{erg/cm^2}$ at room temperature for the spacer thickness range $0.5<d^{\mathrm{MgO}}<1$ nm.

Figure 1 shows the temperature dependence of the parallel FMR spectra (saturation signals) for a representative sample $\mathrm{Fe}_{32}/\mathrm{MgO}_{0.57}/\mathrm{Fe}_{3.5}/\mathrm{Co}_{35}/\mathrm{V}_{10}$. Subscripts mark the corresponding layer thicknesses in nanometers. The optical mode is observed at higher fields than the acoustic one implying an AF coupling.[14] In the inset, the intensity ratio of the modes is plotted as the function of the temperature. Such behavior of optical and acoustic modes and of their intensity ratio denotes a weakening of the coupling with the temperature decrease.

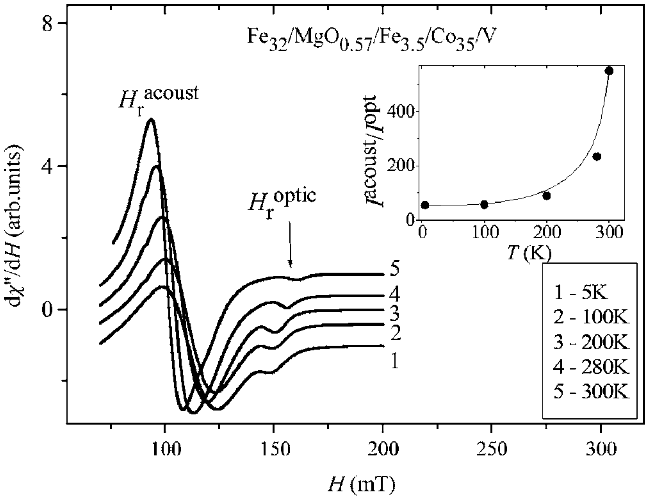

FIG. 1. FMR spectra of a representative $\mathrm{MgO}/\mathrm{Fe}_{32}/\mathrm{MgO}_{0.57}/\mathrm{Fe}_{3.5}/\mathrm{Co}_{35}/\mathrm{V}_{10}$ MTJ at different temperatures. The intensity ratio of acoustic and optical modes, plotted as a function of temperature, is shown in the inset. The spectra were recorded in the parallel configuration of FMR.

Temperature dependencies of the acoustic and optical parallel resonance fields in $\mathrm{Fe}_{34}/\mathrm{MgO}_{0.6}/\mathrm{Co}_{35}/\mathrm{V}_{10}$ and $\mathrm{Fe}_{32}/\mathrm{MgO}_{0.57}/\mathrm{Fe}_{3.5}/\mathrm{Co}_{35}/\mathrm{V}_{10}$ films are shown in Fig. 2. In both stacks, the difference in resonance fields of the two modes increases with increasing temperature. For the sake of clarity, this is also shown in the inset to this figure. The change of $|\delta|$ amounts to ~36% and clearly designates the increase of the AF coupling strength at higher T or vice versa, weakening of the IEC under sample cooling. The sensitivity of the FMR technique to the changes in coupling strength is demonstrated in the inset of Fig. 2 by plotting δ vs d^{MgO} (upper-right scale). The line through the symbols (spheres) represents an exponential fit. Note that in this case, the layer magnetizations and anisotropy fields did not change experimentally.

It is well known that in ferromagnetic films, a dominating inhomogeneous part of the FMR linewidth (ΔH) is mainly sensitive to the anisotropy dispersion and the magnetostriction, both being temperature dependent quantities. The dispersion should be larger at interfaces due to imperfections and strains which tend to relax with increasing temperature. A number of interface regions is greater in stacks than in reference samples; thus, the former are expected to have broader lines. However, in spite of this expectation, it has been found that ΔH in the stacks is essentially smaller than the linewidths in MgO/Co/V and MgO/Fe/Co/V samples mimicking electrodes of MTJs. With layers similar to reference films in magnetic and crystalline structure, the stacks may basically have an additional origin of line narrowing, associated with the interlayer coupling. Could IEC narrowing be a phenomenon similar to a well-known exchange narrowing effect? This issue remains open until an advanced theoretical study is conducted. As regards experiments, a strong correlation between ΔH and IEC has been found.

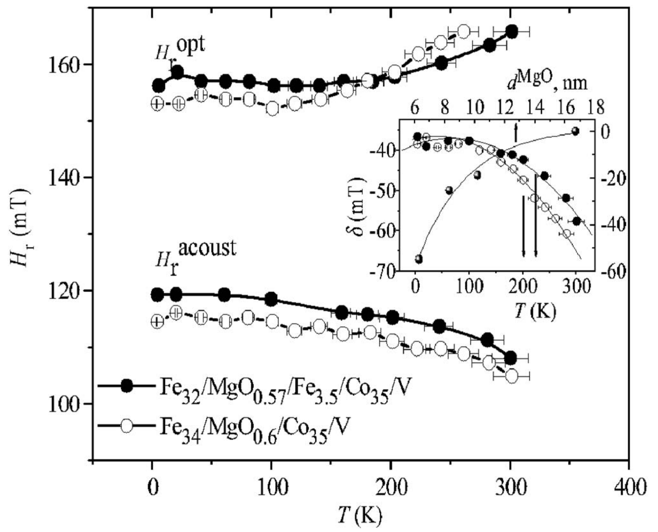

FIG. 2. Temperature dependencies of the acoustic and optical parallel resonance fields are plotted for the $MgO/Fe_{34}/MgO_{0.6}/Co_{35}/V_{10}$ and $MgO/Fe_{32}/MgO_{0.57}/Fe_{3.5}/Co_{35}/V_{10}$ MTJs. The inset depicts the temperature dependencies of $\delta = H_r^{\mathrm{acoust}} - H_r^{\mathrm{opt}}$ in both samples (lower and left scale) and the MgO room temperature thickness dependence of δ in the $MgO/Fe_{35}/MgO_x/Fe_{3.5}/Co_{35}/V_{10}$ MTJ (upper and right scale).

Temperature dependence of the normalized linewidth ($\Delta H/\Delta H_{T=5\ \mathrm{K}}$) in both reference samples and MTJs is depicted in Fig. 3(a) with the dc field applied in plane. On one hand, in MTJs ΔH narrowing under sample heating gets stronger, especially close to the room temperature. As shown by temperature dependencies of resonance fields and signal intensities (Figs. 1 and 2), the IEC is the strongest at 300 K. On the other hand, the FMR linewidth narrowing with IEC increase was also found in MTJs at room temperature. This is obvious in Fig. 3(b) where ΔH is plotted either as a function of the difference between the resonance fields of acoustic and optical modes (curve δ) or depending on the exchange field obtained using SQUID (curve H_{ex}). In both cases, the films were magnetized in the plane. The growth of $|\delta|$ and $|H_{\mathrm{ex}}|$ implies IEC increasing.

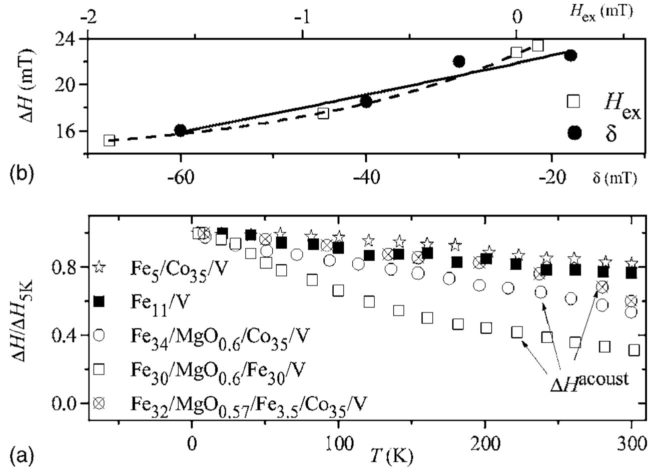

FIG. 3. (a) Temperature dependencies of the normalized linewidth $\Delta H/\Delta H_{\mathrm{at}\ T=5\ \mathrm{K}}$ in MTJs and in reference films. (b) Dependencies of the FMR linewidth on the distance between acoustic and optical modes (curve δ) and on the exchange field (curve H_{ex}) in the MTJs $Fe_{35}/MgO_x/Fe_{3.5}/Co_{35}/V_{10}$ at $T=300$ K. Lines are exponential fits. The dc field is applied in the film plane.

In summary, temperature dependencies of X-band FMR fields, signal intensities, and linewidths in epitaxial MTJ-systems Fe/MgO/Co/V and Fe1/MgO/Fe2/Co/V, as well as in reference films, have been studied. FMR data indicate the AF interlayer coupling occurring at very thin MgO barriers (<1 nm). The sign of the coupling correlates with a shift of a minor hysteresis loop. The temperature behavior of FMR fields and signal intensities in the range of 2–300 K allow concluding that the coupling strength increases with heating. This is in agreement with a pure tunneling mechanism and its more sophisticated version taking into account the interfacial realistic electronic structure.[7–10] To this point, noteworthy are the results obtained on our Fe/MgO/Fe films, where shot noise analysis demonstrates a pure tunneling mechanism and absence of defects within the MgO barrier.[17] All adduced experimental results invalidate theoretical approaches promoting the resonant assisted tunneling mechanism related to defects (oxygen vacancies) within the barrier as an origin of the IEC.

Finally, in the parallel FMR configuration under sample heating from 2 to 300 K, the narrowest lines were observed close to the room temperature, where the IEC has been found to be the strongest. At 300 K, ΔH also decreased with increasing IEC while the spacer thickness was reduced. These facts point out an additional narrowing mechanism associated with the exchange coupling. We obtained similar results studying Fe1/MgO/Fe2/V films;[15] however, in MTJs including the Co layer, they are more pronounced.

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