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Testing epitaxial Co_{1.5}Fe_{1.5}Ge(001) electrodes in MgO-based magnetic tunnel junctions

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The ability of the full Heusler alloy Co_{1.5}Fe_{1.5}Ge(001) (CFG) to be a Half-Metallic Magnetic (HMM) material is investigated. Epitaxial CFG(001) layers were grown by molecular beam epitaxy. The results obtained using electron diffraction, X-ray diffraction, and X-ray magnetic circular dichroism are consistent with the full Heusler structure. The pseudo-gap in the minority spin density of state typical in HMM is examined using spin-resolved photoemission. Interestingly, the spin polarization found to be negative at E_F in equimolar CoFe(001) is observed to shift to positive values when inserting Ge in CoFe. However, no pseudo-gap is found at the Fermi level, even if moderate magnetization and low Gilbert damping are observed as expected in HMM materials. Magneto-transport properties in MgO-based magnetic tunnel junctions using CFG electrodes are investigated via spin and symmetry resolved photoemission.

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The material science community is putting a large effort into developing original magnetic materials for the implementation in low-energy spintronic devices. An important part of this work is devoted to growing electrode materials for Magnetic Tunnel Junctions (MTJs) for Magnetic Random Access Memory (MRAM), oscillators, and sensors. Thin magnetic electrodes having high spin-polarization, low damping, moderate magnetization, and high perpendicular magnetic anisotropy are investigated. The family of Half-Metallic Magnetic (HMM) compounds fulfill these requirements as ab initio calculations performed on many Heusler or Half-Heusler compounds (NiMnSb, Co_{2}FeSi, Co_{2}MnSi…) have predicted a 100% spin polarization at the Fermi level. The Curie temperature of Co_{2}FeGe has been measured close to 1000 K, and perpendicular anisotropy for iron rich Co_{20}Fe_{50}Ge_{30} has been demonstrated when sandwiched between MgO layers due to the perpendicular interface anisotropy induced by MgO. Therefore, all these features make CFG a promising candidate for magnetic-RAM electrode. However, CFG has not been successfully integrated in MTJ’s so far and its spin-polarization at and below the Fermi level has not yet been measured.

In this Letter, we report on experimental studies of single crystal Co_{1.5}Fe_{1.5}Ge(001) layers as single films or as electrodes in MgO-based MTJ’s. First, the growth and structural characterization of CFG on MgO(001) substrates is discussed. Gilbert damping and magnetic features are extracted from FMR and magnetometry measurements. The spin-polarization of occupied electronic states is determined using spin-resolved photoemission spectroscopy at the SOLEIL synchrotron source. Finally, transport measurements performed on CFG/MgO(CFG(001) and Co_{25}Fe_{75}/MgO/CFG(001) MTJs are presented.

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Single CFG films were grown by molecular beam epitaxy (base pressure $10^{-10}$ Torr) on MgO(001) single-crystal substrates. The CFG single layers were grown by co-evaporating Co, Fe, and Ge using 3 Knudsen cells whose fluxes were calibrated by using a quartz microbalance located at the sample position. The uncertainty on the concentrations is estimated to be $\pm 5\%$. The films were deposited on MgO at room temperature, and annealing temperatures were tested in the range of 350°C–700°C. Three types of MTJs were grown on MgO(001) substrates. Here, we will mostly discuss MTJ’s with structure: Fe (20 nm)/CFG (8 nm)/Co (20 nm)/Au (10 nm) and Co$_{53}$Fe$_{75}$ (50 nm)/MgO (2.4 nm)/CFG (30 nm)/Co (20 nm)/Au (10 nm). The CFG electrodes were deposited at room temperature and then annealed during 30 min at temperatures between 350°C and 500°C. The whole stacking was subsequently patterned by UV-photolithography and a conventional physical etching processes to get MTJ cells with a junction’s size of 10 × 10 to 50 × 50 $\mu m^2$.

The structural properties of single CFG films were first analysed during growth using Reflection High Energy Electron Diffraction (RHEED). At room temperature, the RHEED patterns are the same as observed when growing pure Fe films (Fig. 1). The unit cell is, thus, a square of half the size of the Heusler (001) face, meaning that no chemical ordering occurs. Annealing at 250°C is sufficient to observe new 1/2 streaks appearing along the (11) azimuth of the initial square lattice (Fig. 1), meaning that the lattice cell has doubled in size as expected for the Heusler L2$_1$ structure. It should be noted that this superstructure cannot be observed in the case of the full Heusler X$_2$YZ structure with chemical disorder between Y and Z (B2 phase). In the case of the X$_{1.7}$Y$_{1.2}$Z phase, however, a more complex chemical arrangement cannot be ruled out. This simple observation is a clear indication that some chemical ordering takes place even at low annealing temperatures $<300$°C.

To go further, X-ray diffraction experiments were performed using a Cu K$_{\alpha 1}$ anode ($\lambda = 0.154$ nm) along the (001) direction. Fig. 1 shows the typical X-ray $\theta$–$2\theta$ diffraction pattern for an annealed CFG film. In addition to the MgO (002) peak (not shown), the pattern shows only the (002) and (004) peaks typical of the Heusler structure. No additional phase (e.g., a parasitic hexagonal phase) has been detected. The lattice spacing is equal to 0.573 nm in agreement with values already reported. The full determination of the species distribution within the lattice is not possible here because the Co and Fe scattering factors are very close using K$_{\alpha 1}$ X-rays. However, it is possible to get some information on the location of Ge atoms by fitting the (002) and (004) peaks. A Rietveld refinement was used and an excellent agreement is obtained (simulated curves in Fig. 1) assuming the full Heusler L2$_1$ structure. To go further simulations were performed assuming the substitution of some Fe or Co atoms by Ge. We thus were not able to fit the experimental data. However, this simulation does not allow us to eliminate the possibility of some chemical disorder between Fe and Co atoms. Such Co and Fe permuation is predicted to destroy the pseudogap.

The magnetization is also very sensitive to chemical disorder. We performed magnetometry experiments using a commercial VSM with automatic sample rotation. The magnetization at saturation on average is 1000 ± 100 kA/m (emu/cm$^3$) equivalent to 5 ± 0.5 $\mu m$ per unit cell, which is similar to calculated values for Co$_{53}$FeGe$^6$ and experimental values in Ref. 4. A cubic magnetic anisotropy is deduced from the hysteresis loop shape, especially from the value of remanent magnetization as a function of the applied magnetic field angle. X-ray magnetic circular dichroism (XMCD) was also performed on the DEIMOS beamline at SOLEIL (Fig. 2(a)) at the Fe, Co, and Ge 2p edges. A dichroic signal is observed for the 3 elements. The atomic magnetic moments of Fe and Co were found parallel, and antiparallel to the Ge one. The application of the sum rules (using the Fe and Co number of holes in the bulk) gives 2.0 ± 0.1 $\mu B$/at and 1.5 ± 0.1 $\mu B$/at for Fe and Co, respectively. The Ge magnetic moment is difficult to determine but is very small (small dichroic signal). Theoretical calculations based on first principles within density-functional theory using the plane-wave ultrasoft pseudo-potential method within the generalized gradient approximation for the exchange-correlation functional$^{11,12}$ have been performed to determine the magnetic moments. To study the effect of permutations we have used a 16 atom super cell. The fully relaxed structure has a total magnetic moment of

![FIG. 1. RHEED patterns obtained on a CFG film as-deposited at RT (top left) and annealed (top right). New 1/2 streaks appear after annealing typical of chemical ordering. The (001) diffraction peak intensities are also well fitted assuming the full Heusler L2$_1$ structure. (bottom).](image)
spin DOS at the Fermi level. All these results are strong indications that chemical disorder is very limited in our films.

The CFG spin polarization was investigated using Spin-Resolved PhotoEmission Spectroscopy (SR-PES) performed on the CASSIOPEE beamline at the SOLEIL synchrotron. Single CFG films were epitaxial grown in a molecular beam epitaxy (MBE) chamber connected to the beamline\textsuperscript{15} so that surface contamination is prevented. First SR-PES experiments were conducted with 37 eV \( p \) polarized light at a 45\(^{\circ} \) incident angle. The photoelectron detection was performed along the (001) normal axis of the films using the largest aperture (+/−8\(^{\circ} \)) leading to investigating 80\% of the Brillouin Zone (BZ). The spin resolution was achieved by using a Mott detector. The sample temperature was maintained below 100 K during the experiments. Figures 3(a) and 3(b) show the majority \( N_{\uparrow} \) and minority \( N_{\downarrow} \) photoemission spectra (PES) obtained on single CoFe (as reference layer) and single CFG thin films annealed at around 550°C. The corresponding spin polarizations \( P = (N_{\uparrow} - N_{\downarrow})/(N_{\uparrow} + N_{\downarrow}) \) are plotted in Figs. 3(a) and 3(b). In the reference CoFe layer, the expected negative spin polarization is found at \( E_F \) (Fig. 3(a)). The situation is clearly different for CFG films. The main difference between CoFe and CFG consists in a strong majority spin contribution around \( −0.2 \text{ eV} \) leading to a positive polarization peak at the same energy value (Fig. 3(b)). This new majority spin band pushes the spin polarization to become close to \( E_F \). The occurrence of this strong majority spin contribution may be at the origin of the greatly enhanced magnetoresistive properties obtained in CFG spin-valves compared to CoFe\textsuperscript{5}. However as might be expected our experiment on CFG spin-valves films shows that the actual spin polarization is <100%.

A final test to evaluate HMM behavior is to use CFG as electrodes in MgO-based MTJs. A first experiment was to grow CFG on Fe/MgO(001) underlayers since the resulting MgO barrier quality is excellent and well understood in this case. The upper electrode was a 10 nm thick CFG layer covered by a thick Co layer resulting in a magnetic coercive

\begin{center}
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\end{center}

FIG. 1. Typical XAS and XMCD signal on Co and Fe for CoFe(001) and (b) (CoFe)\textsubscript{3}Ge films.  

\begin{center}
\includegraphics[width=\textwidth]{figure2.png}
\end{center}

FIG. 2. Magnetic properties of (CoFe)\textsubscript{3}Ge film using X-Ray absorption spectroscopy (XAS and XMCD) and FMR. (a) The XMCD signal on Ge is opposite to the Fe and Co signals, suggesting that the Ge magnetic moment is coupled antiparallel to Fe and Co. (b) FMR frequency versus perpendicularly applied field. The red line is a linear fit, yielding a Landé factor of 2.07 and a magnetization of 1.25 T. Inset: Polar magneto-optical Kerr effect loop (10 nm film). (c) FMR linewidth versus frequency in perpendicular applied field conditions (30 nm film). Inset: Real and imaginary parts of the permeability for a field of 2 Tesla.

\begin{center}
\includegraphics[width=\textwidth]{figure3.png}
\end{center}

FIG. 3. PES spectra (top) and spin polarization (bottom) for annealed (a) equimolar CoFe(001) and (b) (CoFe)\textsubscript{3}Ge films.
field for the upper electrode substantially higher than that of the lower Fe electrode. The CFG layer was grown at RT and annealed up to 550 °C. The Co layer was grown on top of CFG at RT to avoid intermixing. The Tunnel Magneto-Resistance (TMR) vs. H curves are shown in Fig. 4. The TMR reaches 125% at RT and more than 220% at 15 K consistent with a good epitaxial process and high spin polarization in the tunneling process. However, these values are smaller than state of the art TMR values obtained on Fe/ MgO/Fe(001) around 200% at RT and 450% at 10 K, see Ref. 16. The TMR is even lower using two CFG electrodes (Fig. 4), where the TMR is only around 15% at RT and 60% at 15 K. To better understand these magneto-transport results, we carefully studied the symmetry of the electronic states near EF.

To summarize, single crystalline Co$_{1.5}$Fe$_{1.5}$Ge (001) films were grown by molecular beam epitaxy. The structure of the films is cubic with the expected lattice constant. All the structural and magnetic characterizations clearly indicate chemical ordering consistent with the full Heusler structure. In particular, very low Gilbert damping coefficients are obtained. However, some chemical disorder involving Co atoms occupying in Y sites instead of Fe cannot be ruled out here, which would lead in accordance to our theoretical investigations to a suppression or reduction of the pseudo-gap at the Fermi level. The spin-polarization of CFG(001) at EF is observed to be positive opposed to the negative spin-polarization of FeCo(001), and, furthermore it is not 100%. The pseudo-gap for minority spin at EF is not observed. Some minority spin DOS with Δ$_1$ symmetry was observed at the Fermi energy explaining the modest TMR values obtained in MgO-based MTJs using CFG electrodes.

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