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Negative magnetoresistance in Fe_3O_4/Au/Fe spin valves

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The structural, electrical, and magnetic properties of Fe_3O_4/Au/Fe spin valves on MgO(001) are presented. In contrast to more conventional spin valve structures, the current-in-plane resistance of the Fe_3O_4/Au/Fe spin valves is found to be smallest for an antiparallel alignment of the magnetization of the Fe_3O_4 and Fe layer. Since the electrical current is transported through the low resistance Au and Fe layers, the negative magnetoresistance effect is attributed to an inverse electron spin scattering asymmetry at the Fe_3O_4/Au interface.

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The incorporation of Fe_3O_4 films into magnetoresistance (MR) devices has attracted much scientific attention in recent years. The main reason for this is its high Curie temperature of 858 K and the prediction that Fe_3O_4 exhibits full negative spin polarization at the Fermi level.[1,2] Although experiments have not confirmed the complete half-metallicity of Fe_3O_4, they do show that the number of minority electrons is larger than the number of majority electrons at the Fermi level.[3–7] Since the spin polarization of Fe_3O_4 is opposite to that of most other magnetic materials commonly used in magnetic tunnel junctions (MTJs) and magnetic spin valves, the use of one Fe_3O_4 electrode in these devices is expected to yield negative MR effects, i.e., the electrical resistance is lowest for an antiparallel alignment of the magnetization direction of the two ferromagnetic layers. For MTJs negative tunneling magnetoresistance (TMR) has indeed been observed,[5–7] but until now only positive giant magnetoresistance (GMR) effects have been reported for magnetic spin valves with one Fe_3O_4 layer.[8,9]. In this paper we show that Fe_3O_4/Au/Fe spin valves exhibit negative GMR in the current-in-plane geometry, due to opposite electron spin scattering asymmetries at the Fe_3O_4/Au spacer layer interface and in the Fe layer and at the Fe/Au interface.

The Fe_3O_4/Au/Fe spin valves were grown by dc-magnetron sputtering on MgO(001) substrates in a Leybold Z550-S system with a base pressure of 10^{-7} mbar. The Fe_3O_4 films were reactively sputtered from a pure Fe target in 3×10^{-3} mbar Ar and 4×10^{-5} mbar O_2 at a substrate temperature of 673 K. Deposition of Fe_3O_4 on MgO(001) under these optimized conditions results in single crystalline films with a (001) orientation. In addition, resistance measurements on these films show a clear Verwey metal-insulator transition around 120 K, which is characteristic for Fe_3O_4 and an indication of good quality film growth.[10,11] The Au and Fe films were grown at substrate temperatures below 373 K. The deposition rates were determined by small angle x-ray diffraction reflectivity measurements. The magnetotransport properties of the spin valves were measured in the van der Pauw configuration. After contacting the 10 mm square samples in the four corners, the samples were placed in an electromagnet with a maximum field of 160 mT. The MR of the spin valves was measured in the longitudinal ($H\parallel I$) and transverse ($H\perp I$) geometry. Superconducting quantum interference device (SQUID) magnetometry was used to study the magnetization reversal processes in the spin valve structures.

Figure 1 shows a $\theta-2\theta$ x-ray diffraction scan of a MgO(001)/30 nm Fe_3O_4/5 nm Au/10 nm Fe/2 nm Au spin valve. Since the lattice parameter of Fe_3O_4 ($a=8.396$ Å) is approximately twice that of the MgO(001) substrate ($a=4.213$ Å), the absence of any distinct Fe_3O_4 reflections in Fig. 1 indirectly indicates the growth of an (001)-oriented Fe_3O_4 film on MgO(001). X-ray diffraction scans of Fe_3O_4 films on $SrTiO_3(001)$ substrates do indeed confirm the growth of epitaxial $Fe_3O_4(001)$ films under the selected deposition conditions. Figure 1 reveals a good MgO(001)$\parallel$Au(111) out-of-plane epitaxial relationship. The absence of any clear Fe reflections indicates that Fe grows on the Au(111) spacer layer without a well-defined crystal orientation. From transmission electron microscopy and small angle x-ray diffraction reflectivity measurements the roughness of the interfaces in the spin valve is estimated to be smaller than 1 nm.

Figure 2 shows the resistance of a 30 nm Fe_3O_4/5 nm Au/10 nm Fe/2 nm Au spin valve as a function of the applied magnetic field in the longitudinal and transverse geometry at 90 and 300 K. Two different MR effects can be distinguished. In small magnetic fields the resistance of the spin valve abruptly decreases in the longitudinal measurement, while it increases in the transverse measurements. This anisotropic magnetoresistance (AMR) is due to magnetization reversal in the Fe layer through which part of the electrical current is transported. In addition to the AMR effect, the

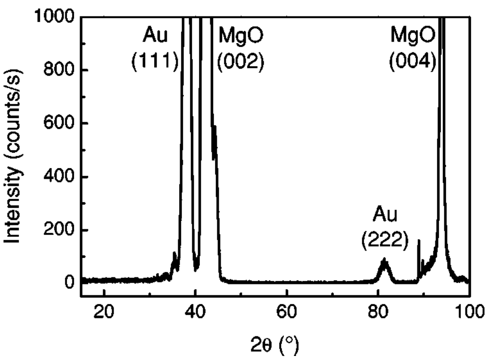

FIG. 1. X-ray diffraction scan of a 30 nm Fe_3O_4/5 nm Au/10 nm Fe/2 nm Au spin valve on a MgO(001) substrate.

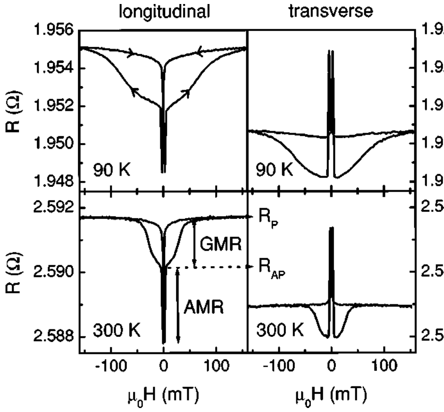

FIG. 2. Longitudinal and transverse magnetotransport measurements on a 30 nm Fe_3O_4/5 nm Au/10 nm Fe/2 nm Au spin valve structure at 90 and 300 K. The arrows indicate the AMR and GMR effects.

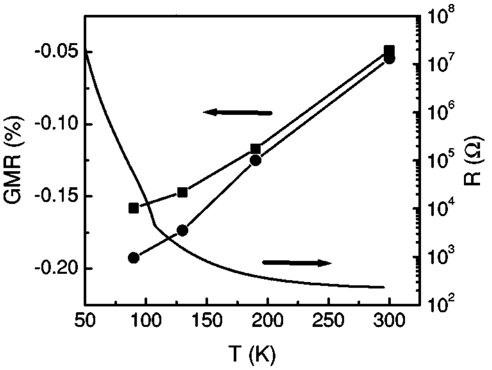

FIG. 4. Temperature dependence of the negative GMR effect in a 30 nm Fe_3O_4/5 nm Au/10 nm Fe/2 nm Au spin valve (squares) and a 15 nm Fe_3O_4/4 nm Au/5 nm Fe/2 nm Au spin valve (circles) and the temperature dependence of the resistance of a 50 nm thick Fe_3O_4 film on MgO(001) (solid line).

Fe_3O_4/Au/Fe/Au spin valve exhibits a clear isotropic GMR. The GMR effect is a result of independent magnetization reversal in the Fe_3O_4 and Fe layers (see Fig. 3). In large magnetic field the magnetization of the two ferromagnetic layers are aligned parallel. When the magnetic field is reversed the magnetization of the Fe layer switches first, which results in a nearly antiparallel alignment of the magnetization direction of the Fe_3O_4 and Fe layer. As can be seen in Fig. 2, the change from a parallel to an antiparallel magnetization configuration leads to a *reduction* of the sample resistance. Finally, the higher-resistance parallel configuration is reestablished by a gradual magnetization reversal in the Fe_3O_4 layer at larger applied magnetic field.

The transport measurements of Fig. 2 show that the resistance of the Fe_3O_4/Au/Fe/Au spin valve is smallest for an antiparallel alignment of the magnetization direction of the Fe_3O_4 and Fe layer. This is opposite to the GMR of more conventional spin valves and the GMR of Fe_3O_4/Cu/Fe (Ref. 8) and Fe_3O_4/Au/NiFe (Ref. 9) trilayers. To extract the GMR values from the transport measurements we define $\mathrm{GMR}=(R_{\mathrm{AP}}-R_{\mathrm{p}})/R_{\mathrm{p}}$ and we average over the longitudinal and transverse measurements to cancel out the AMR contributions (see Fig. 2). Using these definitions the negative GMR of the 30 nm Fe_3O_4/5 nm Au/10 nm Fe/2 nm Au spin valve yields −0.16% at 90 K and −0.05% at 300 K.

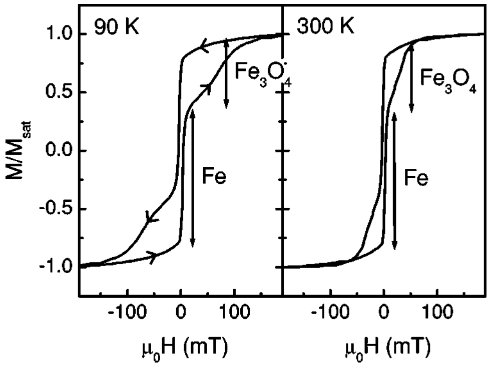

FIG. 3. Normalized SQUID magnetization curves of a 30 nm Fe_3O_4/5 nm Au/10 nm Fe/2 nm Au spin valve on a MgO(001) substrate at 90 and 300 K.

The temperature dependence of the GMR effect for two different Fe_3O_4/Au/Fe/Au spin valves is shown in Fig. 4. The magnitude of the GMR effect increases monotonically with decreasing temperature, which we attribute to reduced spin-flip scattering in the spin valve layers at low temperature. The GMR versus temperature data does not show any correlation with the temperature dependence of the Fe_3O_4 layer resistance, which is shown for a 50 nm thick Fe_3O_4 film in Fig. 4. This is a clear indication that the observed GMR is due to spin-dependent electron scattering at the Fe_3O_4/Au interface instead of electron scattering in the Fe_3O_4 layer. The shunting of the in-plane electrical current by the metallic Au and Fe layers is confirmed by a comparison of the resistance of the different spin valve layers. For the 30 nm Fe_3O_4/5 nm Au/10 nm Fe/2 nm Au spin valve, the resistance of the Fe_3O_4 layer at 300 K is 383 Ω (from Fig. 4). This is much larger than the resistance of the entire spin-valve structure of about 2.5 Ω (see Fig. 2). The difference in resistance between the Fe_3O_4 layer and the Au/Fe/Au trilayer increases rapidly with decreasing temperature. At 90 K the resistance of the 30 nm Fe_3O_4 layer is about 1×10^5 Ω, while the resistance of the spin valve is only 1.9 Ω. From these resistance values the maximum GMR effect that can possibly originate from electron scattering in the Fe_3O_4 layer is estimated to be −0.0006%. This value, which is obtained when $(\rho\downarrow/\rho\uparrow)_{\mathrm{Fe_3O_4}}=0$ (full half-metal) and $(\rho\downarrow/\rho\uparrow)_{\mathrm{Fe}}=2$ are used in the two-current GMR model,[12] is more than two orders of magnitude smaller than the experimentally measured GMR effect.

Negative GMR effects have previously been measured on Fe/Cu multilayers with thin Cr layers within half of the Fe layers,[13] FeV/Au/Co spin valves,[14] and Co/Ru/Co trilayers.[15] The negative GMR of these structures is explained by an inverse spin scattering asymmetry ($\alpha=\rho\downarrow/\rho\uparrow$) in the ferromagnetic layers or at their interfaces, i.e., $\alpha(F1)>1$ and $\alpha(F2)<1$ or vice versa. If the magnetization of the two magnetic layers is antiparallel, the weakly scattered majority electrons in one of the layers are weakly scattered minority electrons in the other layer. This leads to a shunting of the spin valve current by a low resistance spin channel in the antiparallel configuration and hence $\rho_{AP} < \rho_P$ (negative GMR).

To explain the negative sign of the GMR effect in the Fe_3O_4/Au/Fe/Au spin valves the electron scattering asymmetry at the Fe_3O_4/Au interface must be opposite to that in the Fe layer and at the Fe/Au interfaces. Since $\alpha_{Fe} > 1$, this means that majority electrons are scattered more than minority electrons at the Fe_3O_4/Au interface ($\alpha_{Fe_3O_4/Au} < 1$). The interface scattering asymmetry might be due to electron scattering on magnetic ions at the interface. In this scenario, conduction electrons probe the interfacial layers of the Fe_3O_4 film. Although the large bulk resistance significantly limits the transport of electrons within the Fe_3O_4 film, there are no obvious reasons why electrons are not able to probe the interfacial layers with a certain penetration depth. An alternative explanation for the electron scattering asymmetry is a ferromagnetic proximity effect between the Fe_3O_4 film and the Au interfacial layers. Contact between ferromagnetic Fe_3O_4 and nonmagnetic Au can lead to an induced magnetic moment on the Au atoms near the Fe_3O_4/Au interface. As a consequence, the low resistance Au layer will exhibit a small spin scattering asymmetry in the vicinity of the Fe_3O_4/Au interface and together with the spin scattering asymmetry in the Fe layer and at the Fe/Au interfaces it will contribute to a GMR effect. The negative sign of the GMR effect can be explained by $\alpha_{Au} < 1$, i.e., the resistivity of minority electrons is lower than the resistivity of majority electrons. Hence, the induced spin scattering asymmetry in the Au interfacial layers will have the same polarity as the spin scattering asymmetry in Fe_3O_4: In both cases the electrical current is dominated by minority electrons.

The existence of a ferromagnetic proximity effect between the Fe_3O_4 and Au layer of a Fe_3O_4/Au/Fe/Au spin valve is not unlikely. *Ab initio* calculations using relativistic local spin density theory show that the average induced spin and orbital magnetic moments on Au interfacial atoms in a Fe/Au multilayer is about 0.07 μ_B and 0.03 μ_B, respectively.[16] In addition, induced magnetic moments on 5*d* elements such as W, Ir, and Pt have been measured with x-ray magnetic circular dichroism.[17] These measurements show that the total induced magnetic moment in Fe/W and Fe/Ir multilayers amounts about 0.2 μ_B per W and Ir atom.

Negative MR effects have also been measured on magnetic tunnel junctions with one Fe_3O_4 electrode and a $La_{0.7}Sr_{0.3}Mn_{0.3}$ counterelectrode, in which the negative TMR is due to tunneling or hopping of electrons between two electrodes with opposite spin polarization.[5,6] In the Fe_3O_4/Au/Fe/Au spin valves the negative GMR is due to opposite electron spin scattering asymmetries at the Fe_3O_4/Au spacer layer interface and in the Fe layer and at the Fe/Au interface. The negative GMR in our spin valves is opposite to the positive GMR in Fe_3O_4/Cu/Fe trilayers (measured in the current-in-plane geometry)[8] and Fe_3O_4/Au/NiFe spin valves (measured in the current-out-of-plane geometry).[9] Possible explanations for this discrepancy are the quality of the Fe_3O_4/metal interface, the influence of the spacer layer material (Cu versus Au), and the difference in the measurement geometry. For measurements in the current-out-of-plane geometry, the current is forced to pass through the Fe_3O_4 layer. Consequently, an intrinsic negative MR from the Fe_3O_4 layer is also measured, which complicates the determination of a small GMR effect.

In summary, we have successfully demonstrated that spin valves with one Fe_3O_4 layer can exhibit negative GMR. Current-in-plane magnetotransport measurements on Fe_3O_4/Au/Fe/Au spin valves indicate that the resistance is lowest for an antiparallel alignment of the magnetization of the Fe_3O_4 and Fe layer. The negative GMR is −0.05% at 300 K and its magnitude increases monotonically with decreasing temperature. At 90 K the GMR yields −0.16% for a 30 nm Fe_3O_4/5 nm Au/10 nm Fe/2 nm Au and −0.19% for a 15 nm Fe_3O_4/4 nm Au/5 nm Fe/2 nm Au spin valve. Since the high resistance Fe_3O_4 layer does not transport a significant amount of the electrical current, the negative GMR effect is attributed to an inverse spin scattering asymmetry at or in the vicinity of the Fe_3O_4/Au interface. At this interface majority electrons are scattered more than minority electrons.

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