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Suppression of spin transport in ferromagnet/oxide/semiconductor junctions by inserting magnetic impurities

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Abstract
We have investigated the effect of magnetic impurities in a tunnel barrier on the spin transport in ferromagnet/oxide/semiconductor (FM/I/SC) junctions. Fe/GeO_2 tunnel contacts were fabricated on Ge substrates with submonolayer amounts of Mn intentionally incorporated in the middle of the GeO_2 tunnel barrier. Spin transport was probed using the three-terminal (3T) Hanle geometry at 15 K. Control junctions without Mn impurities exhibit typical Hanle signals with features characteristic of spin accumulation and spin precession. However, the magnitude of the spin voltages decreases with increasing amount of Mn dopants and the spin signals are completely absent for the devices having 0.2 nm of Mn impurities. This demonstrates that magnetic impurities in a tunnel barrier are detrimental to the spin transport in magnetic tunnel contacts on a semiconductor.

1. Introduction

The integration of the spin functionality in the conventional complementary-metal-oxide semiconductor technology is intended to develop a new generation of devices that combine high performances and very low power consumption. To realize spin-based devices with semiconductors, the transfer of spin information from a ferromagnet (FM) into a semiconductor (SC) is essential. Over the last decade, several research groups have succeeded to electrically induce and probe a spin polarization at room temperature in SCs using different device structures and measurement configurations [1-5]. Among them, the 3T scheme with either a FM/oxide tunnel contact [1-4] or a direct Schottky contact of a ferromagnetic metal [5,6] on a SC has been particularly widespread thanks to its simple implementation and robustness. However, the origin of the spin signals observed in 3T devices with oxide tunnel barriers has been heavily debated because the measured spin signals are usually orders of magnitude larger [1,2,4] than what the standard theory predicts [7]. Alternative theories based on two-step tunneling and spin accumulation in localized states or impurity-assisted tunneling magnetoresistance have therefore been proposed [2,8-9]. These theories involve the presence of localized states, either in the oxide or at the oxide/semiconductor interface. Surprisingly, very large spin accumulation signals were recently observed in a direct Schottky contact of ferromagnetic Mn_5Ge_3 on Ge, without an oxide tunnel barrier [6]. Therefore, the enhanced spin signal cannot be explained by any mechanism that involves the oxide and the localized states these may produce. In this experiment, it is possible, in principle, that some Mn atoms might have diffused into the Ge during the growth process and produced deep levels in the depletion region of the Ge. One may then wonder whether or not two-step tunneling via these deep levels might have enhanced the spin signal. Although some previous studies have demonstrated that the presence of impurities in magnetic tunnel junctions leads to a strong suppression of the tunnel magnetoresistance and is therefore detrimental for the spin transport [10,11], the effect of magnetic impurities on the Hanle spin signals in ferromagnetic tunnel contacts on semiconductors has not yet been investigated.

In this study, we have intentionally inserted Mn impurities in the middle of an oxide tunnel barrier and investigated the effect on the spin transport in FM/I/SC junctions. We demonstrate that the incorporation of submonolayer amounts of Mn impurities in the barrier suppresses the Hanle spin signal. This proves that magnetic impurities are detrimental to the spin transport and thus cannot be the cause of the large Hanle signals observed in FM/I/SC devices.

2. Experimental details

Ferromagnetic tunnel contacts were grown at room temperature by molecular beam epitaxy on heavily doped p-type Ge(001) substrates. The tunnel barrier was prepared as follows: first the Ge substrate was covered with a 0.65 nm-thick GeO_2 oxide layer by electron-beam evaporation. Subsequently, an amount of Mn impurities varying from 0.05 to 0.2 nm was evaporated using an effusion cell while the chamber was cooled with liquid nitrogen to minimize surface diffusion and clustering effects. After this step, a GeO_2 layer with a thickness varying from 0.65 to 1 nm was deposited under oxygen atmosphere (~ 1×10^{-6} Torr). Finally, a 5 nm-thick Fe layer and Au cap layer were evaporated. To confirm the layer thicknesses, the crystalline quality and the composition profile of the junctions, cross-sectional observations and elemental analysis were carried out using a transmission electron microscope (TEM) equipped with energy dispersive X-ray spectroscopy and high-angle annular dark field imaging in scanning mode (HAADF-STEM) operating at 200 kV. For the transport measurements, junctions were prepared with standard micro-fabrication techniques. The details of the device structures, dimensions and junction-resistance area products (junction-*RA*) are summarized in Table I. Hanle measurements were performed at 15 K using the 3T configuration. The reproducibility of the observed Hanle signals was verified by measuring several tunnel junctions on the same sample and all the results were compared to that of a simultaneously prepared control sample without Mn impurities labeled device A.

Table I Details of the device structures, dimensions and junction-*RA*s used in this study.

Device	Mn thickness (nm)	GeO_2 thickness (nm)	Junction dimension (μm × μm)	Junction-*RA* at 15 K and 100 mV (Ωμm²)
A	0	1.3	200 × 100	5.71×10^5
B	0.05	1.3	100 × 50	1.05×10^5
C	0.1	1,3	100 × 50	3.97×10^5
D	0.2	1.7	100 × 50	3.42×10^5

3. Results and discussion

The high-resolution cross-sectional TEM images of the control sample (device A) and of the sample containing 0.2 nm of Mn impurities (device D) are presented in Fig. 1. No substantial differences are visible between the two junctions. The GeO_2 oxide barriers and the Fe layers are amorphous and polycrystalline, respectively. The interfaces with the Ge have a peak to valley roughness lower than 0.3 nm for both devices. More importantly, no clusters or inclusions are observable in the Mn-doped junction. The presence and spatial distribution of the Mn dopants were then investigated by HAADF-STEM observations. The observations (not shown here) clearly reveal the presence of Mn in the oxide region, and show no evidence that the Mn atoms have segregated or diffused into the electrodes. Hence, all these results demonstrate that the Mn dopants were distributed within the GeO_2 and that the FM/oxide tunnel contacts used in this study have a comparable crystalline quality.

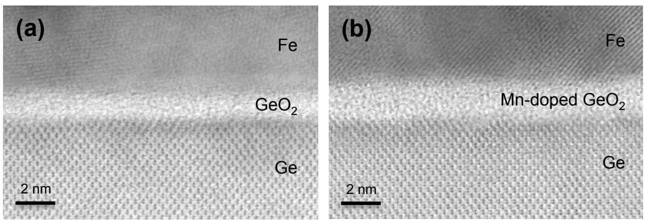

Fig. 1 High-resolution cross-sectional TEM images of the $Fe/GeO_2/Ge$ devices (a) without Mn (device A) and (b) with 0.2 nm of Mn impurities (device D).

As shown in Table I, the junction-*RA*s vary by less than a factor of 5, allowing a fair comparison between all the devices. In addition, all the junctions exhibit nonlinear and almost symmetric current-voltage characteristics.

The Fig. 2 presents the Hanle curves measured at 15 K at a constant current and when a magnetic field perpendicular to the film plane ($B_\perp$) is applied. For the devices A, B and C, the spin voltages obtained show all the characteristic features of an induced non-equilibrium spin population: a signal decay with a Lorentzian shape for small $B_\perp$ due to spin precession, then an increase of the signal for larger $B_\perp$ due to rotation of the ferromagnet towards the out-of plane direction. Interestingly, the magnitude of the Hanle signal gradually decreases with increasing amount of Mn impurities. The spin signal is even completely absent for the device D. Therefore, the presence of Mn impurities in the tunnel barrier clearly suppresses the Hanle spin signal. Taking into account that the Mn ions have a magnetic moment, the strong reduction of the Hanle signal is most likely due to inelastic spin exchange scattering of the tunneling electrons with the magnetic ions. We thus conclude that magnetic impurities are detrimental to the spin transport and thus cannot be the cause of the large Hanle signals observed in FM/I/SC devices.

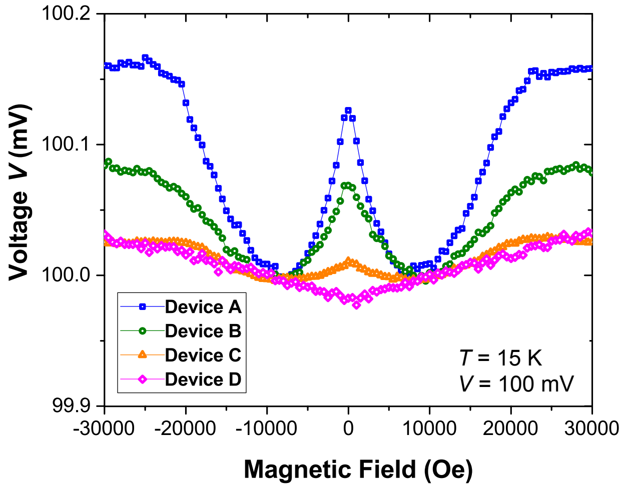

Fig. 2 Hanle spin signals of the $Fe/GeO_2/Ge$ devices having different amounts of Mn impurities. All the measurements were performed at 15 K and at a bias voltage V = 100 mV. A linear background was subtracted from the data.

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