Electrical detection of spin accumulation in a p-type GaAs quantum well.

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We report on experiments in which a spin-polarized current is injected from a GaMnAs ferromagnetic electrode into a GaAs quantum well through an AlAs barrier. The resulting spin polarization in the GaAs well is detected by measuring how the current, tunneling to a second GaMnAs ferromagnetic electrode, depends on the orientation of its magnetization. Our results can be accounted for the non-relaxed spin splitting of the chemical potential, that is spin accumulation, in the GaAs well. We discuss the conditions on the hole spin relaxation time in GaAs that are required to obtain the large effects we observe.

Introducing the spin as an additional degree of freedom in semiconductor devices is an important challenge for the future of spintronics\textsuperscript{1,2}. The semiconductors combine the advantage of a long spin lifetime with the flexibility of their carrier concentration and their high mobility. The long spin coherence time in semiconductors has been evidenced by time-resolved optical experiments and, for example, a spin lifetime reaching a fraction of $\mu$s has been evidenced in n-doped GaAs at low temperature\textsuperscript{3,4}. However, the prerequisite of spin injection from a ferromagnetic conductor in most concepts of devices raises difficult problems. It has turned out that injecting spins from a ferromagnetic metal encounters difficulties related to the conductivity mismatch between metal and semiconductor\textsuperscript{5,6} and also to their possible chemical incompatibility. This has driven the development of magnetic semiconductors\textsuperscript{7-9}, like Ga$_{1-x}$Mn$_x$As which is ferromagnetic up to 110 K\textsuperscript{8}, more adapted for integration into semiconductor heterostructures. Successful experiments on spin injection have been achieved by injecting an electrical current from magnetic semiconductors or metals and detecting the circular polarization of emitted light\textsuperscript{10-14}. However, if the principle is now well established, there is some questions on how the efficiency of spin injection can be extracted from the light polarization\textsuperscript{15}. In this letter we present experiments of spin injection from a GaMnAs electrode into a GaAs quantum well with detection of the polarization in GaAs by measuring how the current, tunneling from GaAs into a second GaMnAs electrode, depends on the orientation of its magnetic moment. The structure is a double tunnel junction GaMnAs/AlAs/GaAs/AlAs/GaMnAs. The first junction plays the role of ballistic spin injector whereas the second one is used to detect the spin accumulated in the semiconductor before being transmitted. Our observation of large tunnel magnetoresistance (TMR) effects demonstrates the efficient spin transmission across GaAs. This is in contrast with the absence of spin transmission in double junctions when the base is a nonmagnetic metal, and can be explained by the non-relaxed spin polarization predicted for a semiconductor base\textsuperscript{6}.

Our double tunnel junctions, grown by molecular beam epitaxy on semi-insulating GaAs (001), are composed of two ferromagnetic electrodes (Ga$_{1-x}$Mn$_x$As) separated by a AlAs(1.5nm)/GaAs(5nm)/AlAs(1.5nm) trilayer. Thin layers of GaAs (1nm) are also intercalated between the GaMnAs and AlAs layers to prevent interdiffusion between the two materials. To probe the spin-polarization of electron tunneling from GaMnAs through AlAs, test experiments have been also performed on single tunnel junctions where the central trilayer of the double junction is replaced by a single 1.7 nm thick AlAs barrier\textsuperscript{16}. Structures have been deposited at 230°C on a GaAs buffer layer grown at 580°C. Junctions with diameter from 10$\mu$m to 300$\mu$m were patterned by optical lithography\textsuperscript{17}. Ohmic contacts on both GaMnAs electrodes were made by deposition of Ti (50nm) and Au (150nm).

Different thicknesses and Mn concentrations have been chosen for the two electrodes in order to obtain different coercive fields and then an antiparallel magnetic configuration. The bottom and top Ga$_{1-x}$Mn$_x$As films have respective thicknesses of 300nm and 30nm. The Mn concentration is 4.3% and 5.3% (bottom and top electrode) for the double barrier structure, 4.7% and 5.4% (bottom and top) for the single barrier. M(H) hysteresis loops of the heterostructures before patterning show two steps associated to the reversal of the two GaMnAs layers at different coercive fields. The remanent magnetization is only 30% of the saturated magnetization which is reached at about 1 Tesla. The magnetization of the sample collapses near 50 K (Curie temperature) and the absence of remanent magnetization above this temperature indicates there is no formation of MnAs clusters. Higher Curie temperatures than 50K have been obtained for Ga$_{1-x}$Mn$_x$As with $x \approx 5\%$ after thermal treatments. Nevertheless, we have not annealed our junctions to avoid
possible diffusions into the AlAs barriers. We have however checked the TMR of our single junctions (a probe of the spin polarization) is nearly as high (38%) as for the junctions with the same AlAs thickness in Ref.[9].

In Fig.1, we show the TMR of the double barrier (Fig.1a) and single barrier (Fig.1b) junctions at 4 K. In both cases, the magnetic field is set along the [100] magnetic easy axis and the TMR is derived from four-contact measurements at constant bias voltage (1 mV). The TMR ($\Delta R/R_0$ where $R_0$ is the zero field resistance) is associated with the switching between the parallel (P) and antiparallel (AP) configurations of the remanent magnetizations (30% of saturation). Similar TMR results on single barrier junctions have been found by Tanaka and Higo. For a thickness of 1.7 nm for AlAs, these authors find a TMR ratio $\cong 45\%$ that is approximately the same as what we measure on single barrier structure.

Figure 2 shows that the TMR amplitude decreases rapidly with the bias voltage. This bias dependence of the TMR is derived from the difference between I(V) curves (inset of Fig.2) recorded at zero field and in the antiparallel magnetic configuration. The bias dependence of Fig.2 is confirmed by R(H) curves recorded at different bias.

A striking result is that the double junctions exhibit TMR effects (at a level similar to that of the single junction), in contrast to which could be expected for F/I/N and N/I/F junction in series. In such F/I/N/I/F double barrier where N is nonmagnetic, TMR is expected in the following cases:

a) Hot electrons having not relaxed their energy and transmitted above the second barrier$^{19,20}$. This can produce a significant TMR for applied voltage exceeding the barrier height but not in the very small voltage limit of our experiments.

b) Predominant direct tunneling between the ferromagnetic electrodes through the entire AlAs/GaAs/AlAs barrier would give TMR effects but also a much higher tunnel resistance. According to the results of Tanaka and Higo, increasing the thickness of AlAs from 1.7 nm (thickness in our single barrier junction) to 3.4 nm (double barrier) would increase the resistance by almost four orders of magnitude and, in addition, decrease the TMR. We can rule out direct tunneling in our double junctions because their resistance and TMR are both close to those of the single junction (around $10^{-2}\Omega.cm^2$ for the resistance and 38% for the TMR).

c) Coherent resonant tunneling on quantum well states in GaAs would give TMR effects but would be characterized by a specific bias dependence of the conductance (a negative differential conductance for example)$^{22}$ and TMR. We have plotted on Fig.2 the bias dependence of the TMR and conductance in inset. We do not observe any fine structure that could be the signature of a coherent resonance on discrete levels. Actually, a coherent resonant tunneling would require a coherence time of the wave functions in the well, $\tau_c$, longer than the mean time spent by the hole in the well, $\tau_n$. This condition will be discussed later.

d) Sequential tunneling without spin relaxation in the GaAs well (or more generally in a semiconducting spacer) can give TMR effects, as calculated in Ref. For this process is expected to produce a large TMR if the spin relaxation time $\tau_{sf}$ is larger than the mean time $\tau_n$ spent ballistically in the spacer between the two successive tunnelings (in principle, the TMR of the double junctions should be half that of the simple ones, but, as our double junctions have slightly thinner AlAs barriers, the increase of the spin-polarization of tunneling at decreasing thickness$^{9}$ should balance more or less the reduction by a factor of 2). For the situation of diffusive transport in the spacer, this corresponds to the second condition of Eq.35 in Ref.6, that is $\tau_r \ll \rho N t_{sf}^2/\tau_n$ or equivalently $t_N \ll \rho N t_{sf}^2/\tau_T$ ($t_N$=spacer thickness, $\rho N$ and...
\( l_e^N \) = resistivity and spin diffusion length in the spacer, \( r_T \) = tunnel resistance). To our knowledge, such sequential tunneling with spin conservation has never been observed up to now.

We ascribe the TMR of our double junction to the mechanism d). This is also supported by the discussion below on the three characteristic times \( \tau_n \), \( \tau_c \) and \( \tau_{sf} \).

The time \( \tau_n \) is related to the broadening of the quantized energy level \( \epsilon_n \) and can be expressed as a function of \( \epsilon_n \) and \( T \) - the transmission coefficient of the detection tunnel barrier - by \( \tau_n = \pi\hbar/(\epsilon_n T) \). This expression can be directly derived from the picture of holes reflecting 2/\( T \) times against the barriers with a kinetic energy \( \epsilon_n \) before being ejected out of the well. A typical energy of some tens of meV for a few nm thick well results in a value of \( \tau_n \) of the order of 100 ps for a transmission coefficient \( T \) of the order of \( 10^{-3} \) (this is the value derived from the variation of the tunnel resistance as a function of the barrier thickness in the experimental results of Tanaka and Higo).

With a value of the order of 100 ps, \( \tau_n \) is much longer than the coherence time \( \tau_c \) (\( \approx \) inelastic relaxation time, which is generally less than 1 ps). This shows that the condition for resonant tunneling is not fulfilled and this implies that we are in a regime of sequential tunneling. In this regime, we expect that the double junction exhibits a TMR at the level of the TMR of the single junction if the spin relaxation time \( \tau_{sf} \) is much longer than \( \tau_n \), that is 100 ps and thus approaches the ns range. This seems to be in agreement with the results of optical measurements on hole spin lifetime in GaAs quantum well. This enhancement of the hole spin lifetime at low temperature compared to the bulk value, measured recently at \( \approx 100 \text{fs} \) \( ^{20} \), can be understood as the effect of i) the lift of the valence-band degeneracy between the \( J_z = \pm 3/2 \) and \( J_z = \pm 1/2 \) states at the \( \Gamma \) point produced by the confinement or equivalently ii) the strong reduction of the solid angle of hole wave-vectors around the quantized direction (that is a small \( k \) parallel component) as \( k_B T \) remains small compared with \( \epsilon_n^{25} \).

The condition \( \tau_{sf} \ll \tau_n \) of the discussion above can be related to the condition expressed in the model of Ref.\(^{6} \), that is in a picture with a splitting of the spin up and spin down electro-chemical potentials (Fermi energies) in the AP configuration (as illustrated in the inset of Fig.1a). This splitting simply reflects that, in the AP configuration, one injects a majority of spin up holes whereas a majority of spin down holes tunnel towards the outer electrode, thus generating an imbalance between the TMR at the level of the order of the total voltage drop \( V \) between ferromagnetic electrodes), this gives rise to the same TMR as in the single junctions. The condition of negligible relaxation is having a number of spin flips per unit of time and unit area in the well, that is \( (\Delta \mu/k_B T)n^{2D}/\tau_{sf} \) at small bias, much smaller than the injected spin current of the order of \( j/e (n^{2D} \) is the density of the 2-Dimensional gas -2DEG- in the well)\(^{6} \). Expressing \( \tau_{sf} \) as a function of the spin diffusion length \( l_{sf} \) in the well and hole mobility \( \nu \) from the relation \( l_{sf} = \sqrt{k_B T \tau_{sf} / e^2} \), the condition for maintaining \( \Delta \mu \) at a level of the order of \( eV \approx \nu r_T J / (r_T \text{ is the tunnel resistance}) \) can be written as :

\[
\tau_T \ll \frac{l_{sf}^2}{n^{2D} e V} \tag{1}
\]

which is the 2DEG version of the condition \( \tau_T \ll \rho_N N_e \bar{\tau}^{2D} / t_N \) in Ref.\(^{6} \). The equivalence of Eq.1 with the condition \( \tau_n \ll \tau_{sf} \) turns out straightforwardly if the tunnel resistance is related to the transmission coefficient \( T \) by a Landauer-like formula, \( r_T = \alpha \hbar / e^2 n^{-1} T^{-128,29} \) and then to \( \tau_n \) by \( \tau_n = \pi\hbar / (\epsilon_n T) \) (the coefficient \( \alpha = k_B T / \epsilon_n \) is a reduction factor expressing that the energy \( \epsilon_n \) of the confined state is larger than \( k_B T \)).

To probe our interpretation, we have also measured the TMR of a double junction with the same value of \( t_N \) (GaAs spacer thickness) but with a higher value of \( r_T \) (by about a factor of 10). The TMR curves are very similar to those of Fig.1 but the amplitude is only 3% instead of 38%. This is consistent with the reduction of the spin-polarization when \( r_T \) becomes too large to satisfy Eq.1. In other words, this means that, with the smaller transmission coefficient \( T \) associated to a higher \( r_T \), the time spent by the hole in the GaAs well, \( \tau_n = \pi\hbar / (\epsilon_n T) \), is no longer much smaller than the spin relaxation time \( \tau_{sf} \) and the spin relaxation in GaAs reduces the TMR.

The spin splitting of the electro-chemical potential in a nonmagnetic spacer between two tunnel junctions has already been detected by Jedema et al.\(^{30} \), this time for a metallic spacer (Cu) in Co/Al\(_2\)O\(_3\)/Cu/Al\(_2\)O\(_3\)/Co double junction structures. However, in this case, the splitting is much smaller than the potential drop between the magnetic electrodes, typically 10 \( \mu \)eV compared to the potential drop of the order of 100 meV. This can be expected from Eq.1 or the equivalent condition for a 3D-spacer, \( r_T \ll \rho^{3D} l_{sf}^2 / t_N \), where \( \rho^{3D} \) is the resistivity of
the spacer. Actually, with the typical low-resistivity of metals (low compared to semiconductors) and spin diffusion length $l_{sf}$ in the micron range, the above condition for obtaining a spin splitting of the order of the potential drop across the double junction would require to have $r_f$ not higher than $0.1 \Omega \mu m^2$. With resistances of alumina barriers of the order of $1k\Omega \mu m^2$, the spin splitting turns out to be a very small fraction of the total potential drop across the double junction. More generally, this also explains that a significant TMR could never be observed in double junctions in which the central layer is a nonmagnetic metal with such high tunnel resistance. The situation with a semiconductor central layer of high resistance is much more favorable.

In conclusion, we have presented experiments in which, after injection of a spin-polarized current into a GaAs quantum well from a GaMnAs electrode, the spin polarization in GaAs is detected by measuring the spin polarization of the current tunneling into a second GaMnAs electrode. We have shown that our results can be explained by sequential tunneling with low enough spin relaxation in the GaAs layer. The TMR of our double junction (38%) has the same order of magnitude as the TMR of the single ones single ones, what can be expected if the condition of negligible relaxation in GaAs, Eq.1, is satisfied. We have also shown that this condition can be hardly satisfied with a metallic spacer instead of GaAs, so that the effects we observe are specific of spin injection into semiconductors. To our knowledge, these experimental results represent the first clear evidence of an electrical spin detection of spin injection into a semiconductor. Further experiments on similar structures with various thicknesses of the central well (or layer) or various dopings and carrier densities should lead to a more general understanding of the conditions for spin injection and electrical spin detection in semiconductors. We point out that injection into n-type semiconductors having a larger spin lifetime should allow spin propagation on longer distances. Injecting spins from a third contact for an additional control of the spin polarization should lead to new types of spintronic devices.

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