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 layer through an AlAs barrier. The resulting spin polarization in GaAs is detected by measuring how the tunneling current, to a second GaMnAs ferromagnetic electrode, depends on the orientation of its magnetization. Our results can be accounted for by sequential tunneling with the nonrelaxed spin splitting of the chemical potential, that is, spin accumulation, in GaAs. We discuss the conditions on the hole spin relaxation time in GaAs that are required to obtain the large effects we observe.

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Introducing the spin as an additional degree of freedom in semiconductor devices is an important challenge for the future of spintronics [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 demonstrated by time-resolved optical experiments and, for example, a spin lifetime reaching a fraction of μs has been evidenced in n-doped GaAs at low temperature [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 [5,6] and also to their possible chemical incompatibility. This has driven the development of magnetic semiconductors [7–9], such as $Ga_{1-x}Mn_xAs$, which is ferromagnetic up to 140 K [10] and 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 [11–15].

In this Letter we present experiments of spin injection from a GaMnAs electrode into a GaAs layer with detection of the polarization in GaAs by measuring how the tunneling current 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 nonrelaxed spin polarization [16] predicted for a semiconductor base [6].

Our double tunnel junctions, grown by molecular beam epitaxy on semi-insulating GaAs (001), are composed of two ferromagnetic electrodes ($Ga_{1-x}Mn_xAs$) separated by a AlAs(1.5 nm)/GaAs(5 nm)/AlAs(1.5 nm) trilayer. Thin layers of GaAs (1 nm) 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 also been performed on single tunnel junctions where the central trilayer of the double junction is replaced by a single 1.7 nm thick AlAs barrier [17]. Structures have been deposited at 230 °C on a GaAs buffer layer grown at 580 °C. Junctions with diameter from 10 to 300 μm were patterned by optical lithography. Different thicknesses and Mn concentrations have been chosen for the two electrodes in order to obtain different coercive fields and thus an antiparallel magnetic configuration. The bottom and top $Ga_{1-x}Mn_xAs$ films have respective thicknesses of 300 and 30 nm. The Mn concentration is 4.3% and 5.3% (bottom and top electrode) for the double barrier structure, and 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 30% of the saturated magnetization which is reached at about 1 T. 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. Curie temperatures higher than 50 K have been obtained after thermal treatments for $Ga_{1-x}Mn_xAs$ with $x \approx 5\%$. Nevertheless, we have not annealed our junctions to avoid possible diffusions into the AlAs barriers. We have, however, checked that 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(a) we show the TMR of the double barrier and in Fig. 1(b) the single barrier 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 and antiparallel (AP) configurations of the remanent magnetizations. Similar TMR results on single barrier junctions have been found by Tanaka and Higo [9]. For a thickness of 1.7 nm for AlAs, these authors find a TMR ratio $\cong 45\%$ that is slightly above what we measure on single barrier structure (38%).

The interesting result is the observation of TMR in the double junctions, in contrast with what could be expected for $F/I/N$ and $N/I/F$ junctions in series ($F =$ ferromagnetic, $N =$ nonmagnetic, and $I =$ barrier) and also in contrast with what is found in $F/I/N/I/F$ double junctions when N is a metal. The existence of TMR means that the spin polarization injected into GaAs from one of the GaMnAs electrodes is detected by the second GaMnAs electrode. To our knowledge, this is the first example of *electrical* detection of spin polarization in a semiconductor. To explain this TMR, the choice is between (a) coherent tunneling connecting the two spin-polarized GaMnAs electrodes and (b) sequential tunneling with negligible spin relaxation in the GaAs central layer, as we discuss in the two following paragraphs.

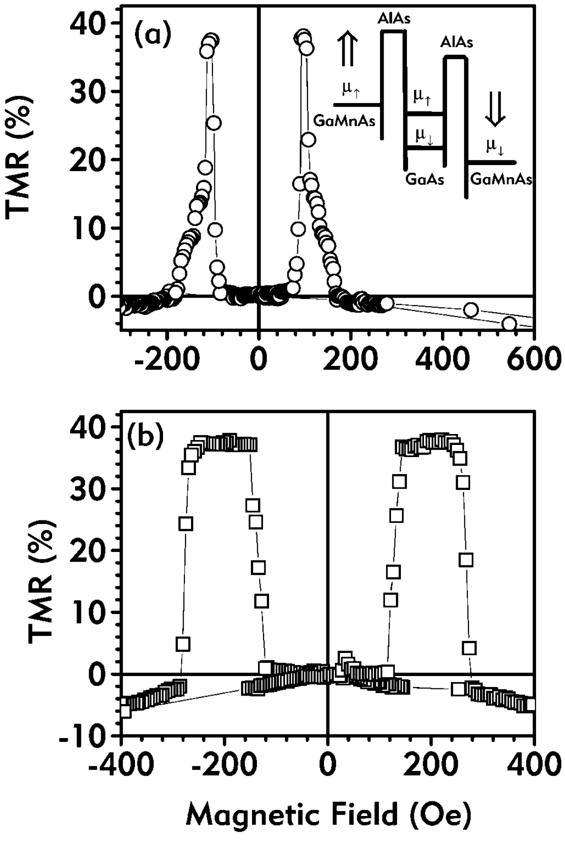

FIG. 1. (a) TMR curve for a 20 μm diameter double barrier junction. Inset of (a): schematic picture of the spin splitting of the electrochemical potentials $\mu_\uparrow$ and $\mu_\downarrow$ in the nonmagnetic central layer of a $F/I/N/I/F$ structure in the antiparallel state (from Ref. [6]). For convenience, the picture for holes has been translated into a picture for electrons. (b) TMR curve for a single barrier MTJ.

(a) Coherent tunneling: We have to distinguish between direct tunneling between the ferromagnetic electrodes through the entire AlAs/GaAs/AlAs barrier and coherent resonant tunneling on quantum well states in GaAs. Direct tunneling through the entire AlAs/GaAs/AlAs barrier would give a much too high junction resistance. According to the results of Tanaka and Higo [9], increasing the thickness of AlAs from 1.7 nm (thickness in our single barrier junction) to 3 nm (total AlAs thickness in our double junctions) would increase the resistance by more than 3 orders of magnitude. We can rule out such direct tunneling because the resistance of the double junctions is close to that of the single barrier junctions (around $10^{-2}\ \Omega\,\mathrm{cm}^2$). In addition, according to Tanaka and Higo [9], the TMR becomes extremely small for AlAs thicknesses above 2.3 nm, whereas we find a large TMR for our double junctions. Coherent resonant tunneling on quantum well states in GaAs would require that the (spatial) coherence time of the wave functions in the GaAs well, τ_ϕ, is longer than the mean time spent by the holes in the well, τ_n. As we show later in the Letter from an estimate of τ_n, this condition is far from being satisfied. It can also be pointed out that we do not observe any of the characteristic bias dependences that are generally associated with resonant tunneling. This can be seen, for example, in the experimental results we present in Fig. 2 for the bias dependence of the TMR and $I(V)$ curve for the double junction.

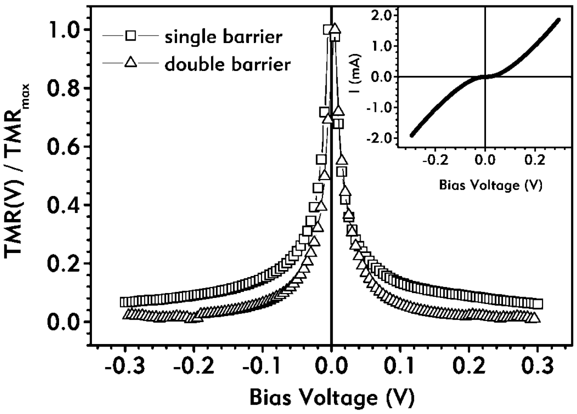

FIG. 2. Bias dependence of TMR for double barrier (triangles) and a single (squares) junctions. Inset: $I(v)$ curve at zero field measured at 4 K on the double barrier junction.

(b) Sequential tunneling without spin relaxation in the GaAs spacer can give large TMR effects if the spin relaxation time τ_{sf} is longer than the mean time τ_n spent by a hole in the spacer between tunneling in and tunneling out. The situation $\tau_{sf} \gg \tau_n$ corresponds to the limit of two independent transport channels for the holes of opposite polarizations throughout the entire structure. A straightforward calculation shows that the TMR of the double junction in this limit is half the TMR of single junctions, $P^2/(1 - P^2)$ instead of $2P^2/(1 - P^2)$ if P is the polarization coefficient of each junction. In our case, as our double junctions have slightly thinner AlAs barriers than our single junctions (1.5 nm instead of 1.7), the increase of TMR from 45% to 70% between 1.7 and 1.5 nm [9,17] should more or less balance the reduction by a factor of 2, and similar values of the TMR can be expected for our single and double junctions in the limit $\tau_{sf} \gg \tau_n$. The following discussion of the respective values of the three characteristic times τ_n, τ_ϕ, and τ_{sf} allows us to ascribe the TMR of our double junctions to the mechanism (b) of sequential tunneling without spin relaxation.

The time τ_n is related to the broadening of the quantized energy level ϵ_n in GaAs and can be expressed as a function of ϵ_n and $\mathbf{T}$ (the transmission coefficient of the detection tunnel barrier) by $\tau_n = \pi\hbar/(\epsilon_n \mathbf{T})$ [18]. This expression can be directly derived in a ballistic picture of holes reflecting $1/\mathbf{T}$ times against the barriers with a kinetic energy ϵ_n before being ejected out of the well. A typical energy of some tens of meV for a few nm thick well [19,20] results in a value of τ_n of a few 100 ps for a transmission coefficient $\mathbf{T}$ smaller than 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 [9]).

With a value larger than 100 ps, τ_n is much longer than the time of phase breakdown τ_ϕ ($\cong$ inelastic relaxation time, which generally does not exceed a few ps at 4 K [21]). This shows that the condition for coherent resonant tunneling is not fulfilled and this implies that we are in a regime of sequential tunneling. As shown above, we expect that our double junction exhibits a TMR at approximately the level of the TMR of the single junction if the spin relaxation time τ_{sf} is much longer than τ_n, that is, longer than 100 ps and thus approaching the ns range. This seems to be in agreement with the results of optical measurements on hole spin lifetime in GaAs quantum well [22]. This enhancement of the hole spin lifetime at low temperature, compared to the bulk value ($\simeq$ 100 fs) in recent measurements [23], 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 Γ point resulting from the confinement or equivalently (ii) the strong reduction of the solid angle of hole wave vectors around the quantization direction (that is, with small parallel components) as $k_B T$ remains small compared with ϵ_n, which slows down the spin relaxation [22].

The condition $\tau_{sf} \gg \tau_n$ 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 electrochemical potentials (Fermi energies) in the AP configuration [as illustrated in the inset of Fig. 1(a)]. 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 (before electrical equilibrium), thus generating an imbalance between the two populations of spin. If the resulting spin splitting $\Delta\mu$ does not relax from its maximum value (of the order of the total voltage drop V between ferromagnetic electrodes), this gives rise to a TMR which is half the TMR of single junctions. The condition of negligible relaxation is having a number of spin flips per unit of time and unit area in the GaAs 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 two-dimensional gas (2DEG) in the well] [6]. Expressing τ_{sf} as a function of the spin diffusion length l_{sf} in the well and hole mobility ν from the relation $l_{sf} = \sqrt{\epsilon_n \nu \tau_{sf}/e}$ [24], the condition for maintaining $\Delta\mu$ at a level of the order of $eV \simeq e r_T J$ (r_T is the tunnel resistance) can be written as

$$r_T \ll \frac{l_{sf}^2}{n^{2D} e \nu}, \tag{1}$$

which is the 2DEG version of the condition $r_T \ll \rho^{3D}[l_{sf}]^2/t_N$ in Ref. [6]. The equivalence of Eq. (1) with the condition $\tau_n \ll \tau_{sf}$ turns out directly if the tunnel resistance is related to the transmission coefficient $\mathbf{T}$ by a Landauer-like formula, $r_T = h/(e^2 n^{2D}\mathbf{T})$ [25,26] and then to τ_n by $\tau_n = \pi\hbar/(\epsilon_n \mathbf{T})$.

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 $\mathbf{T}$ associated with a higher r_T, the time spent by the hole in the GaAs well, $\tau_n = \pi\hbar/(\epsilon_n \mathbf{T})$, is no longer much smaller than the spin relaxation time τ_{sf} and the spin relaxation in GaAs reduces the TMR.

The spin splitting of the electrochemical potential in a nonmagnetic spacer between two tunnel junctions has already been detected by Jedema *et al.* [27], 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 μeV 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, $r_T < \rho^{3D} l_{sf}^2/t_N$, where ρ^{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 that r_T is not higher than 0.1 $\Omega\ \mu\mathrm{m}^2$. With resistances of alumina barriers of the order of 1 $\mathrm{k}\Omega\ \mu\mathrm{m}^2$, 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 been 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.

The last point to discuss, if we consider the bias dependence of the conductance and TMR of our double junctions in Fig. 2, is the absence of any of the features, conductance oscillations as a function of the bias, for example, that can reflect some energy quantization in GaAs. We, however, point out that these features are much less pronounced with p-type semiconductors than in n type. For example, we note that, in the experiments of Ohno *et al.* [20] for an AlAs/GaAs/AlAs quantum well grown at high temperature on GaAs, the amplitude of the oscillations represents only a small fraction of the total conductance. Consequently, it is not surprising to have quite negligible effects in our samples grown on GaMnAs at low temperature [28]. The only manifestation of the quantization in our structures is the long spin relaxation time derived from our experiments.

In conclusion, we have presented experiments in which, after injection of a spin-polarized current into a GaAs layer 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, which can be expected if the condition of negligible relaxation in GaAs, Eq. (1), is satisfied. We have also shown that this condition can not be easily satisfied with a metallic spacer instead of GaAs; as a result, the effects we observe are specific to spin injection into semiconductors. To our knowledge, these experimental results represent the first clear evidence of an electrical spin detection of spin injected 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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[16] For simplicity, although the total angular momentum and not the spin is the good quantum number for the holes in GaAs, we use the term "spin polarization" throughout the Letter. Some differences arising from the distinction between angular momentum and spin will be discussed in a further publication.

[17] 1.7 nm is in agreement with cross-section transmission electron microscopy images of the single junctions. Although the double barriers were grown to obtain the same AlAs thickness, their resistance R is smaller (by a factor of about 2, that is, by a factor of approximately 4 for each individual barrier). The comparison with the $R(t)$ curves of Ref. [9] leads to estimate $t_{\mathrm{AlAs}} \simeq 1.5$ nm in the double junctions.

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