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Resonant tunneling magnetoresistance in MnAs/III-V/MnAs junctions

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This paper investigates the magnetoresistance of micron-sized MnAs/GaAs(AlAs)/MnAs magnetic tunnel junctions. We measure an asymmetric bias dependence of the magnetoresistance in which the negative contribution is attributed to resonant tunneling through a midgap defect band. Within this model we find a spin polarization of 60% for MnAs at the interface with GaAs. Moreover, we show that spin-dependent tunneling is a powerful technique for spectroscopic measurements of defects in a very thin layer.

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One of the main challenges of spintronics today is to combine ferromagnet and semiconductor properties.[1,2] Despite considerable efforts towards room-temperature ferromagnetic semiconductors, the most realistic route to achieve electrical spin injection in semiconductors at room temperature is still to use a ferromagnetic metal combined with a tunnel or Schottky barrier between the metal and the semiconductor.[3–5] One room-temperature ferromagnetic candidate is MnAs,[6] which can be grown epitaxially on standard semiconductor substrates such as Si or GaAs and has abrupt and chemically stable interfaces with III-V semiconductors as AlAs and GaAs.[7] In addition, *ab initio* calculations of MnAs/GaAs/MnAs predict increased spin polarization at the Fermi level for MnAs/GaAs compared with the bulk.[8] Consequently, electrical spin injection and detection with MnAs/(AlAs,GaAs) systems are an important long-term goal.

One way to access the interfacial spin polarization of MnAs/(GaAs,AlAs) systems is to examine the magnetoresistance properties of MnAs/(GaAs,AlAs)/MnAs magnetic tunnel junctions. Assuming direct tunneling between the electrodes, the spin polarization at the Fermi level is related to the tunnel magnetoresistance (TMR).[9] However, small TMR ($<2\%$) has been obtained with MnAs/AlAs/MnAs tunnel junctions[10] and no TMR has been measured through a GaAs barrier.[7] Because GaAs and AlAs layers are grown at low temperature, assisted tunneling through defects has to be considered.[11]

In this paper, we show how a resonant transport picture through a midgap defect band in a GaAs or AlAs barrier can explain the intricate TMR bias dependence measured on a large set of fully epitaxied MnAs/GaAs (AlAs)/MnAs magnetic tunnel junctions of micrometer size. Furthermore, a MnAs/GaAs spin polarization of 60% is obtained when the TMR bias dependence is considered within our resonant tunneling model. This high polarization associated with a relatively low TMR amplitude emphasizes the critical role of defects in achieving spintronic devices. Our results also demonstrate the strong sensitivity of resonant tunneling magnetoresistance as a function of the energy, spatial position, and width of the defect band. Overall, these experiments show that spin-dependent tunneling is a powerful technique for spectroscopic measurements of defects in a very thin layer.

The different tunneling processes of spin-polarized particles can be classified according to their specific spin-dependent conductance $G_{\sigma\sigma'}$. In particular $\sigma,\sigma'=+$ or $-$ where σ (σ') refers to spins emitted (collected) from a ferromagnetic electrode (L) [counterelectrode (R)] and $+$ ($-$) refers to the majority (minority) spin. In the parallel state $\sigma'=\sigma$ whereas in the antiparallel state $\sigma'=-\sigma$ and thus the generalized expression of TMR is

$$\mathcal{T}_{\mathrm{TMR}} = \frac{\int_{\epsilon_{min}}^{\epsilon_{max}} [G_{++}+G_{--}-G_{+-}-G_{-+}]d\epsilon}{\int_{\epsilon_{min}}^{\epsilon_{max}} [G_{+-}+G_{-+}]d\epsilon}. \qquad (1)$$

The processes possible include:

(i) Direct tunneling from the MnAs emitter (L) through the AlAs or GaAs barrier and into the MnAs counterelectrode (R). This gives the standard TMR effect whose amplitude is linked to the spin polarization of the electrodes P_L and P_R at the Fermi level according to the Jullière formula[9]

$$\mathcal{T}_{\mathrm{TMR}} = \frac{2P_LP_R}{1-P_LP_R} \qquad (2)$$

(ii) Impurity-assisted tunneling through a single localized state in the barrier. In this case, the spin-dependent conductance as a function of energy ϵ is given by[12]

$$G_{\sigma\sigma'}(\epsilon-\epsilon_i) = \frac{4e^2}{h}\frac{\Gamma_{L\sigma}\Gamma_{R\sigma'}}{[2(\epsilon-\epsilon_i)]^2+[\Gamma_{L\sigma}+\Gamma_{R\sigma'}]^2}, \qquad (3)$$

where ϵ_i is the energy of the localized state assumed to be not too far from the Fermi level ϵ_F, $\Gamma_{L\sigma}/\hbar$ and $\Gamma_{R\sigma'}/\hbar$ are the leak rates of an electron from the localized state into L and R, respectively. Because the coupling between the electrodes and the impurity is spin dependent, $\Gamma_{L\sigma}=\bar{\Gamma}_L[1+\sigma P_L]$ and $\Gamma_{R\sigma'}=\bar{\Gamma}_R[1+\sigma' P_R]$ are spin-polarized quantities. The overall spin-polarized current is

$$J_{\sigma\sigma'} \propto \int_{\epsilon_F-\beta eV}^{\epsilon_F+(1-\beta)eV} G_{\sigma\sigma'}(\epsilon)d\epsilon, \tag{4}$$

where β describes the spatial position of the impurity in the barrier (such that $\beta=1/2$ when the impurity lies in the center). This leads generally to a strongly asymmetric bias dependence of the TMR. A large negative resonant TMR will result if the impurity is situated asymmetrically in the barrier. Related phenomena observed by Tsymbal *et al.*[13] on Ni/NiO/Co nanojunctions were ascribed to resonant tunneling through a midgap impurity level located randomly in the barrier.

(iii) Single impurity-assisted tunneling through a band or a large number of localized states in the barrier. We assume a large number of localized states situated within a band of width W, centered at a mean energy ϵ_c from ϵ_F and having a Lorentzian energy distribution. The conductance per impurity can be determined by integrating Eq. (3) over this band[14] resulting in

$$G_{\sigma\sigma'}(\epsilon-\epsilon_c) = \frac{4e^2}{h}\frac{\Gamma_{L\sigma}\Gamma_{R\sigma'}}{\Gamma_{L\sigma}+\Gamma_{R\sigma'}} \times \frac{\Gamma_{L\sigma}+\Gamma_{R\sigma'}+W}{[2(\epsilon-\epsilon_c)]^2+[\Gamma_{L\sigma}+\Gamma_{R\sigma'}+W]^2}. \tag{5}$$

The spin-polarized current is given by Eq. (4). From Eq. (5) it follows that the resonance and its associated large negative TMR [case (ii)] is gradually weakened as the bandwidth W becomes comparable to $\Gamma_{L,R}$. There are two important limits here:

(a) $W \gg \Gamma_{L,R}$. In the limit of infinite bandwidth, resonance effects are completely lost. The reminiscent flat-band TMR originating from the envelope factor [first term in Eq. (5)] is positive and equivalent to the sequential TMR of a spin-conserving double tunnel junction with a nonmagnetic central electrode.[5] This TMR, already shown experimentally in p-type GaMnAs/AlAs/GaAs/AlAs/GaMnAs structures,[15] has a positive sign and a magnitude equal to

$$\frac{4\overline{\Gamma_L\Gamma_R}}{[\overline{\Gamma}_L+\overline{\Gamma}_R]^2} \times \frac{P^2}{[1-P^2]}$$

(for identical spin polarization P for L and R), which is strongly dependent on the defect position in the barrier (through the $\Gamma_{L,R}$ parameters).

(b) $W \sim \Gamma_{L,R}$. When the bandwidth W is of the same order of magnitude as the intrinsic level broadening $\Gamma_{L,R}$, the negative resonant tunneling is partly compensated by the positive flat-band envelope contribution. Despite a significant spin polarization of the electrodes, the residual TMR is generally much smaller than the resonant case, whereas its bias dependence remains strongly asymmetric.

(iv) Inelastic tunneling through several localized states. In general, this case leads to a reduction of TMR (Ref. 16) and will not be considered henceforth.

MnAs (100 nm)/AlAs (4 nm)/MnAs (20 nm) and MnAs (100 nm)/GaAs (7.5 nm)/MnAs (30 nm) epilayers were prepared on a GaAs(111)B substrate in a conventional III-V molecular beam epitaxy (MBE) chamber. Thin GaAs layers (1 nm) were inserted at MnAs/AlAs interfaces to avoid Mn diffusion in the AlAs barrier.[17] Details of the low-temperature (LT) growth procedure will be given elsewhere.[14] Samples were then transferred under ultrahigh vacuum to a metallization chamber and capped with gold. Structural characterizations were made *ex situ* with cross-sectional high-resolution transmission electron microscopy (HRTEM). The HRTEM image shows the excellent crystalline quality of the heterostructure (Fig. 1). The planar relation of the system is GaAs(111)B/MnAs(0001)/AlAs(111) [and GaAs(111)]/MnAs(10$\bar{1}$1), perpendicular to the growth direction. GaAs and AlAs barriers were continuous with good crystalline quality and atomically abrupt interfaces (1–2 monolayers). *In situ* x-ray and ultraviolet photoemission study of both MnAs/GaAs and GaAs/MnAs interfaces revealed a GaAs valence band maximum of 0.75±0.1 eV below the Fermi level of MnAs. There was neither apparent band bending (within the scale of ±100 meV) nor interfacial reactivity.[14] From those measurements, we deduced an approximate barrier height of 0.7 eV for MnAs/GaAs. Magnetic tunnel junctions were patterned using an advanced photolithography process allowing sizes as small as 6 μm^2.[18] Magnetotransport measurements were performed with a four-probe method in a continuous He flow cryostat allowing temperature down to 4 K and an in-plane magnetic field up to 7 kOe.

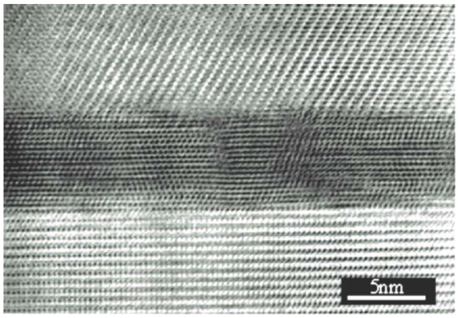

FIG. 1. (Color online) HRTEM cross section of MnAs (100 nm)/AlAs (4 nm)/MnAs (20 nm) from bottom to top with GaAs[1$\bar{1}$0] projection.

Figure 2 shows the magnetotransport measurements at 4 K for an 8-μm^2 cross-section area MnAs/AlAs (4 nm)/MnAs magnetic tunnel junction. The nonlinear dependence of current with dc bias [inset of Fig. 2(b)] is indicative of tunneling.[19] Figure 2(a) displays the magnetoresistance at 10 mV bias. A large variation of resistance is observed corresponding to the reversal of the magnetization of each MnAs layer (50 Oe and 1 kOe) with the applied field in plane along GaAs[1$\bar{1}$0]. Superconducting quantum interference device (SQUID) measurements (not shown here) before patterning were also performed and corroborate the antiparallel configuration of the two MnAs layers in the magnetic field range of 50 Oe to 1 kOe. The magnitude of TMR reaches 12% at 1 mV, which represents the larger value measured on a tunnel junction constituted of two MnAs electrodes. As shown in Fig. 2(b), the bias dependence of the TMR, extracted from $R(H)$ curves, is symmetric and due to magnon excitations decreases monotonically with a characteristic value $V_{1/2}$ of 60 mV.

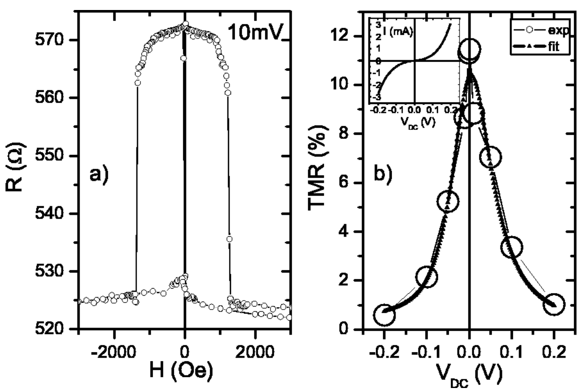

FIG. 2. Data of an 8-μm^2 MnAs/AlAs (4 nm)/MnAs tunnel junction recorded at 4 K. (a) Field dependence of the resistance at 10 mV dc bias showing a positive TMR of 10%. (b) Bias dependence of the TMR deduced from $R(H)$ data (open circles). The solid triangles represent the calculated value with $P=60\%$, $\Gamma_L=7$ meV, $\Gamma_R=57$ meV, $\epsilon_c=-55$ meV, and $W=150$ meV. Inset: $I(V)$ data in the parallel configuration.

Nevertheless, the majority of junctions from the same sample exhibit lower TMR and an asymmetric bias dependence. Such behavior is also demonstrated in MnAs/GaAs (7.5 nm)/MnAs junctions for which we obtain a TMR maximal amplitude of about 2% together with a TMR inversion more or less pronounced at finite bias (from −0.5% to 0%). This behavior was observed for all of the 10 junctions measured on the sample with cross-sectional areas ranged from 8 to 32 μm^2. Typical characteristics are shown in Fig. 3 for a junction of 16 μm^2. Clear asymmetric bias dependence of the TMR is visible with a maximum positive (negative) TMR occurring at 40 mV (−40 mV), Fig. 3(b) [3(c)]. A larger coercivity of the bottom MnAs layer (300 Oe) is observed with this sample, attributed to slight changes in the growth conditions.

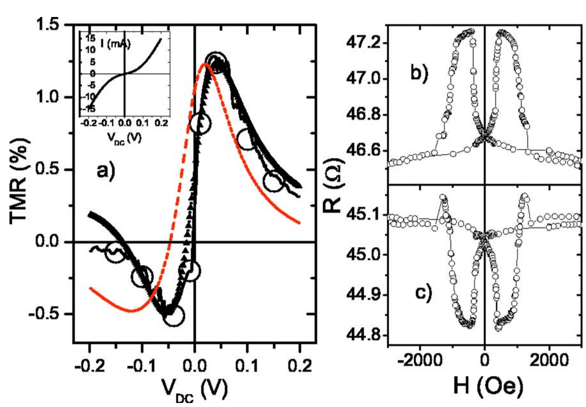

FIG. 3. (Color online) Data of a 16-μm^2 MnAs/GaAs (7.5 nm)/MnAs tunnel junction recorded at 4 K. (a) TMR bias dependence extracted from $I(V)$ (black line) and $R(H)$ (open circles) data. The solid triangles represent the calculated value with $P=60\%$, $\Gamma_L=6$ meV, $\Gamma_R=67$ meV, $\epsilon_c=-40$ meV, and $W=140$ meV. The dashed line corresponds to the calculated value with $P=25\%$, $\Gamma_L=3$ meV, $\Gamma_R=133$ meV, $\epsilon_c=-110$ meV, and $W=70$ meV. Inset: $I(V)$ data in parallel configuration. (b) Field dependence of the junction resistance at 40 mV dc bias. (c) Field dependence of the junction resistance at −40 mV.

Until now, negative TMR and related asymmetric bias dependence were reported and ascribed to (a) direct tunneling between electrodes of opposite spin polarization related to specific interfacial electronic band structures[20,21] or (b) resonant tunneling across a single defect in nanometric-sized junctions.[13] Because of the relative low resistance area (RA) product (~ 1 kΩ μm^2) of our micron-sized junctions given the barrier height (0.7 eV) and thickness (7.5 nm),[22] we ascribe our TMR effects to localized state-assisted tunneling into a broad impurity band described previously as mechanism (iii).

Under this assumption, all the TMR bias dependencies were fitted using Eq. (5) and Eq. (1) with an additional Lorentzian damping function having a characteristic parameter $eV_{1/2}=60$ meV. Experimental data are well reproduced at all positive and negative bias [solid triangles in Fig. 3(a)] by a MnAs polarization P_L and P_R of 60% at each interface, a defect bandwidth W of 140 ± 10 meV and a mean energy ϵ_c ranging from −50 to −40 meV. The latter values agree well with characteristics of the deep donor band appearing in LT-grown GaAs epilayers.[23] Such defects are formed from excess arsenic incorporated primarily as arsenic antisites ($10^{19}-10^{20}$ cm^{-3}) and act as deep donors. Those donors are partly compensated by gallium vacancies (acceptors); as a result the Fermi level is pinned within this deep donor band. This is exactly what we obtain with a Fermi level under the top half part of the defect band with the condition $(\epsilon_F-\epsilon_c)<W/2$. The defect bandwidth is found to be in the same range as those (150−250 meV) extracted from spectroscopic measurements performed on thick GaAs layers.[24,25].

The MnAs spin polarization obtained using this fitting procedure on MnAs/GaAs/MnAs junctions is 60%, much larger than that extracted from the Jullière formula ($P=25\%$) applied to a 12% maximum TMR. As shown on Fig. 3(a) (dashed line), the best fit obtained for $P_L=P_R=25\%$ does not reproduce the experimental data at negative bias. The high polarization associated with a relatively small TMR amplitude originates from the competition between negative resonant TMR and positive contribution from the flat-band envelope [case (iii)], thus reducing the overall TMR amplitude. By contrast, using an identical 60% spin polarization, the bias dependence of TMR for AlAs junctions is also well reproduced [Fig. 2(b)].[26] With a 150-meV broad impurity band, TMR can increase from 1.4% to 12% by reducing the band energy distribution by 15 meV and moving the defect towards the middle of the barrier, thus increasing the positive contribution of TMR. This clearly demonstrates the large sensitivity of spin-dependent tunneling with a defect position (in space and energy).

On the other hand, from the leak rates, $\overline{\Gamma_L}=6$ meV and $\overline{\Gamma_R}=67$ meV, one can estimate the transfer time of electrons in the impurity band to some $\hbar/\bar{\Gamma}\simeq 10^{-14}$ s. This small value demonstrates a transfer through a localized state with high kinetic energy as typically played by deep levels. Finally, we estimate that TMR can exceed 50% in MnAs/GaAs/MnAs systems in the case of a large defect density located near the center of the barrier, that is when $\overline{\Gamma_L}=\overline{\Gamma_R}$. From this ideal case, the introduction of a small asymmetry enhances a negative resonant contribution and thus strongly diminishes the amplitude of TMR.

In conclusion, we measured up to 12% tunnel magnetoresistance with epitaxial MnAs/AlAs (4 nm)/MnAs tunnel junctions, which constitutes the highest TMR measured with two MnAs electrodes. From a large set of measurements with MnAs[/GaAs (1 nm)]/AlAs (4 nm)[/GaAs (1 nm)]/MnAs and MnAs/GaAs (7.5 nm)/MnAs tunnel junctions, we deduced that resonant tunneling occurs through a midgap defect band. Using an appropriate resonant tunneling model including six parameters (two of them, namely the spin polarization and the asymmetry of position, are really unknown; the others can be estimated from literature and measurements), a spin polarization of 60% was assigned to MnAs/GaAs. In addition, we showed that a resonant inversion of the TMR with micrometer-sized tunnel junctions is still possible. Next, we measured the width and the mean position of the impurity band ascribed to the presence of deep levels induced by low temperature excess arsenic in GaAs and AlAs. We have demonstrated the large sensitivity of spin-dependent tunneling with the position and energy of an impurity band and shown its use as a spectroscopic tool in ultrathin layers. Finally, despite the large defect densities in GaAs and AlAs layers, we anticipate a large tunneling magnetoresistance (over 50%), by introducing symmetrically placed defects in the barrier during growth.

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[1] G. A. Prinz, Science **282**, 1660 (1998).
[2] S. A. Wolf, D. D. Awschalom, R. A. Buhrman, J. M. Daughton, S. von Molnàr, M. L. Roukes, A. Y. Chtchelkanova, and D. M. Treger, Science **294**, 1488 (2001).
[3] G. Schmidt, D. Ferrand, L. W. Molenkamp, A. T. Filip, and B. J. van Wees, Phys. Rev. B **62**, R4790 (2000).
[4] E. I. Rashba, Phys. Rev. B **62**, R16267 (2000).
[5] A. Fert and H. Jaffrès, Phys. Rev. B **64**, 184420 (2001).
[6] J. B. Goodenough and J. A. Kafalas, Phys. Rev. **157**, 389 (1967).
[7] M. Tanaka, Semicond. Sci. Technol. **17**, 327 (2002).
[8] K. Narita and M. Shirai, Physica E (Amsterdam) **10**, 433 (2001).
[9] M. Jullière, Phys. Lett. **54A**, 225 (1975).
[10] S. Sugahara and M. Tanaka, J. Cryst. Growth **251**, 317 (2003).
[11] L. I. Glazman and K. A. Matveev, Sov. Phys. JETP **67**, 1276 (1988).
[12] M. Buttiker, IBM J. Res. Dev. **32**, 306 (1988).
[13] E. Y. Tsymbal, A. Sokolov, I. F. Sabirianov, and B. Doudin, Phys. Rev. Lett. **90**, 186602 (2003).
[14] V. Garcia, H. Jaffrès, M. Eddrief, M. Marangolo, V. H. Etgens, and J.-M. George (unpublished).
[15] R. Mattana, J.-M. George, H. Jaffrès, F. N. Van Dau, A. Fert, B. Lépine, A. Guivarc'h, and G. Jézéquel, Phys. Rev. Lett. **90**, 166601 (2003).
[16] R. Mattana, M. Elsen, J.-M. George, H. Jaffrès, F. N. Van Dau, A. Fert, M. F. Wyczisk, J. Olivier, P. Galtier, B. Lépine, A. Guivarc'h, and G. Jézéquel, Phys. Rev. B **71**, 075206 (2005).
[17] M. Tanaka and Y. Higo, Phys. Rev. Lett. **87**, 026602 (2001).
[18] M. Bowen, M. Bibes, A. Barthélémy, J.-P. Contour, A. Anane, Y. Lemaître, and A. Fert, Appl. Phys. Lett. **82**, 233 (2003).
[19] W. F. Brinkman, R. C. Dynes, and J. M. Rowell, J. Appl. Phys. **41**, 1915 (1970).
[20] J. M. De Teresa, A. Barthélémy, A. Fert, J.-P. Contour, F. Montaigne, and P. Seneor, Science **286**, 507 (1999).
[21] C. Tiusan, J. Faure-Vincent, C. Bellouard, M. Hehn, E. Jouguelet, and A. Schuhl, Phys. Rev. Lett. **93**, 106602 (2004).
[22] The corresponding RA product estimated for direct tunneling is $10^8\ \Omega\ \mu m^2$.
[23] M. Stellmacher, R. Bisaro, P. Galtier, J. Nagle, K. Khirouni, and J. C. Bourgoin, Semicond. Sci. Technol. **16**, 440 (2001).
[24] Y. H. Chen, Z. Yang, Z. G. Wang, and R. G. Li, Appl. Phys. Lett. **72**, 1866 (1998).
[25] S. Lodha, D. B. Janes, and N.-P. Chen, J. Appl. Phys. **93**, 2772 (2003).
[26] This is not incompatible to take the same spin polarization for MnAs/GaAs and MnAs/AlAs as thin layers of GaAs are inserted between MnAs and AlAs.