Shot Noise in Epitaxial Double-Barrier Magnetic Tunnel Junctions

Juan Pedro Cascales¹, Laura Martin¹, Amandine Dulluard², Michel Hehn², Coriolan Tiusan^{2,5}, Tomasz Szczepański³, Vitalii Dugaev³, Józef Barnaś⁴, and Farkhad G. Aliev¹

¹Dpto. Física de la Materia Condensada, C03, INC and Condensed Matter Physics Center (IFIMAC), Universidad Autónoma de Madrid, Madrid, 28049 Spain

²Institut Jean Lamour, Nancy-Université Vandoeuvre Les Nancy Cedex, 54506 France

⁵Centre for Superconductivity, Spintronics and Surface Science (C4S), Technical University of Cluj-Napoca, Cluj-Napoca,

400114 Romania

³Department of Physics, Rzeszów University of Technology, Rzeszów, 35-959 Poland

⁴Faculty of Physics, Adam Mickiewicz University, Poznań, 61-614 Poland

We demonstrate that shot noise in Fe/MgO/Fe/MgO/Fe double-barrier magnetic tunnel junctions is determined by the magnetic configuration of the junction—the P-state with the magnetic moments of all three ferromagnetic layers aligned parallel, the AP1 state with the central electrode magnetized opposite to its neighbors, and two different AP2 states with the magnetic moment of the upper or bottom electrode aligned opposite to the other two. We also show that the asymmetry between both MgO barriers is another important factor which affects the shot noise. Some voltages present an enhancement in both conductance and shot noise, which indicates that resonant tunneling through quantum well states formed in the middle magnetic layer is taking place. When the junctions are heated to 60 K, the resonance tunneling anomalies in the shot noise smear out, but they survive in the differential conductance. On the other hand, at low voltages (below 200 mV) and low temperatures (below 4 K) the shot noise tends to decrease, probably due to multi-step tunneling via localized defect states in the tunnel MgO barrier. The theoretical model of sequential tunneling proposed for this system, which takes into account spin relaxation, successfully describes the experimental observations in the bias range between 200 mV and 500 mV, where the influence of tunneling through barrier defects and resonant states inside the central electrode is negligible.

Index Terms—Fano factor, magnetic noise, resonance tunneling devices, shot noise, tunnel magnetoresistance.

I. INTRODUCTION

• HE discovery of huge magnetoresistance effect in Fe/MgO/Fe magnetic tunnel junctions (MTJs) has enormously increased the interest in spin-dependent electron transport in magnetic nanostructures with MgO barriers [1]–[4]. These junctions are actually crucial for most of the modern spintronic devices. Recently, nanostructuring of insulating barriers with single quantum dots [5], arrays of nanoparticles [6], [7], or magnetic layers [8] have been considered as a way to increase control of the tunnel magnetoresistance (TMR) through mechanisms which involve electron interactions and correlations inside the barriers. The latter effects become revealed as resonant tunneling through quantum well states (QWSs) in the linear limit or may create spin/charge accumulation and spin torque oscillations of the magnetically soft middle layer in the non-linear regime [9]. Indeed, Berger [10] predicted a strong reduction of the critical current density needed for spin oscillations by a factor of about six in double-barrier magnetic tunnel junctions (DMTJs) in comparison with MTJs. The reduction of the spin torque threshold has been reported so far only for DMTJs based on magnetic semiconductors [11]. The presence of QWSs in the central layer may further enhance the spin torque efficiency of the DMTJs [12], [13].

Being a consequence of the discrete nature of charge carriers taking part in the non-equilibrium transport of charge, spin, any other kind of matter/energy flow, shot noise (SN) offers a unique tool to investigate correlations and coherency of electron tunneling at the nanoscale, beyond the capabilities of dc electron

Color versions of one or more of the figures in this paper are available online at http://ieeexplore.ieee.org.

Digital Object Identifier 10.1109/TMAG.2013.2243410

transport measurements [14]–[16]. The control of spin diffusion and coherency in spin polarized tunneling remains one of the key challenges limiting the further development of hybrid magnetic nanostructures [10]. Generally speaking, the degree of electron coherency defines whether the real DMTJs are to be considered as single coherent devices or just two decoupled tunnel junctions in series.

In the absence of correlations, shot noise is Poissonian (full shot noise) and is practically independent of frequency (up to the quantum limit), with the noise power given by $S_I = 2eI$ in terms of the average current density I and the electron charge e. The Fano factor $F = S_I/2eI$, which represents the normalized shot noise, can be suppressed (F < 1) [14] or enhanced (F > 1) [17] (even beyond the Poissonian value) by electron interactions.

Spin dependent shot noise in DMTJs has been studied only theoretically in the case of tunneling via quantum dots in the Coulomb blockade [14], Kondo [18], and dynamic spin blockade [19] regimes. The variation of shot noise with the angle between magnetizations of ferromagnetic electrodes has been also analyzed [16]. With a single exception [20], the scope of experimental efforts was, however, limited so far mainly to single barrier MTJs with sequential tunneling via defects [21] or with direct tunneling [22].

Our paper reports a systematic investigation of the electron tunneling statistics in double magnetic tunnel junction devices by measuring shot noise in Fe/MgO/Fe/MgO/Fe DMTJs. Here, we extend our recent report on the control of shot noise via three different magnetic states in DMTJs by demonstrating that: (i) there are generally four different magnetic states with four different values of the shot noise and resistance, (ii) the shot noise in the low bias limit indicates the possible influence of sequential tunneling processes, (iii) the influence of QWSs on the shot noise decreases with temperature and is noticeably absent around 60 K, (iv) conductance anomalies due to QWSs remain nearly unchanged to T = 60 K.

Manuscript received October 31, 2012; revised January 08, 2013; accepted January 22, 2013. Date of current version July 15, 2013. Corresponding author: F. G. Aliev (e-mail: farkhad.aliev@uam.es).



Fig. 1. Normalized magnetization hysteresis of the DMTJs, presenting three different magnetic states.

II. EXPERIMENTAL DETAILS

The tunnel junctions were grown by Molecular Beam Epitaxy (MBE) on MgO (100) substrates under ultra high vacuum (UHV) conditions, typically at a base pressure within the 10^{-11} mbar range. Here we concentrate on electron transport and shot noise measurements in a wide temperature range (between 0.3 K and 60 K) in DMTJs with two slightly different barriers, which have the following structure (numbers in brackets represent thickness in nm): MgO//MgO(10)/Cr(42)/Co(10)/Fe1(5)/ MgO(3)/Fe2(5)/MgO(2.7)/Fe3(10)/Co(30)/Au(10). Fe (100) (bcc structure) is epitaxially grown on MgO (100) (NaCl type crystal) because of their similar lattice parameters (mismatch about 3.7%), with the Fe lattice rotated 45° with respect to the MgO. More details on the growth and characterization of the samples can be found in [20]. Fig. 1 shows the magnetization hysteresis of such a sample. Starting from a positive saturation field, the magnetization reversal takes places in two steps corresponding to two different antiparallel magnetic configurations of the junction's electrodes, as discussed later in the next section.

The junctions patterned by UV lithography have an area of $400 \,\mu m^2$. The noise measurements are based on the cross-correlation method, which removes uncorrelated noise from the amplifiers and the noise of the leads. We take into account the non-linearity of the dynamic conductance while converting the obtained voltage noise into current noise. The experimental setup for conductance and shot noise measurements was described previously in [20]–[22].

III. TUNNELING MAGNETORESISTANCS IN DMTJS WITH ASYMMETRIC BARRIERS

Fig. 2(a), (b) presents the simultaneous measurements of the resistance (solid curves) and shot noise (dots) for a fixed current flowing through the DMTJ with some barrier asymmetry. The observed resistance values and the resistance differences between the four magnetic states reasonably correspond to the structure of the DMTJ (absolute and relative MgO barrier thicknesses and junction area). In the following section we analyze in detail the resistance as a function of the magnetic state of the junction (see Fig. 2).

As long as the two MgO barriers differ only slightly, we are able to observe four different resistive states, instead of the two observed in single barrier MTJs. When the field is swept from the one corresponding to the saturated P state $(\downarrow\downarrow\downarrow\downarrow)$ (the arrows indicate orientations of the magnetic moment of all electrodes) to the opposite orientation, the magnetic configuration first changes from the P state (lowest resistance) to the AP1



Fig. 2. Shot noise and resistance at a fixed current during a magnetic field sweep at T = 0.3 K (a) from positive to negative values and (b) from the AP2₁ at negative fields back to positive fields.

 $(\downarrow\uparrow\downarrow)$ state (highest resistance). This is because the central electrode is magnetically the softest one. Further increase of the magnetic field on the negative field side results in a switching to the AP2₁ state $(\uparrow\uparrow\downarrow\downarrow)$ with the resistance between the values corresponding to the P and AP states. When, at this point, the magnetic field is turned back to the positive direction, one can switch the device (before the P state is reached) to a different AP2₂ state $(\uparrow\downarrow\downarrow\downarrow)$ with antiparallel magnetic alignment of the layers adjacent to the thicker MgO barrier.

IV. MAGNETIC FIELD DEPENDENCE OF THE FANO FACTOR IN DMTJS

Let us consider now the dependence of shot noise and resistance on the magnetic state of DMTJs. Figs. 2(a) and 1(b) compare the tunnel resistance of various magnetic states to the corresponding Fano factors. We note that the Fano factor was determined only for fields where the magnetic states are well defined, i.e., outside the transition regions. One clearly observes that: (i) the Fano factor is suppressed below the Poissonian value (F < 1), and (ii) the Fano factor substantially (well outside the error bars) depends on the magnetic state.

As Fig. 2 shows for T = 0.3 K, the current (not the voltage) was kept constant during the experiments. Below, we discuss the details of the bias dependence of the shot noise and conductance in the temperature range from 0.3 K to 60 K. As we shall demonstrate below, the bias dependence of the Fano factor could be influenced at low enough biases and temperatures by the presence of quantum well states in the central (magnetically soft) layer and by possible sequential tunneling through defects inside the MgO barrier.

V. BIAS DEPENDENCE OF THE FANO FACTOR AND CONDUCTANCE

Fig. 3 presents the bias dependence of the Fano factor in the four different magnetic states at low temperatures (Fig. 3(a)), variation of the bias dependence of the Fano factor with temperature (Fig. 3(b)), and the bias dependence of the deviation of the differential conductance from a parabolic background (Fig. 3(c)). The analysis of the conductance anomalies in the



Fig. 3. (a) Dependence of shot noise with voltage for four different magnetic states. (b) Shot noise in the P state at 0.3 K, 4 K and 60 K. The effect of quantum well states is gradually suppressed with increasing temperature. (c) The oscillations in conductance due to QWSs are not affected by the temperature. The curves have been offset for better observation.

wide temperature interval is expected to provide information on the influence of quantum well states in the middle layer on the electron tunneling.

As can be seen in Fig. 3(a), the range between $\pm (0.2-0.5)$ V shows constant values of the Fano factor for all four magnetic states. These values will be used later for fitting the experimental data to the theory. The Fano factor is estimated by measuring the shot noise at each voltage for all states, which corresponds to the frequency independent part of the noise spectrum. From the shot noise values at very low biases, we extrapolated the noise at zero voltage, which we subtracted from all shot noise measurements as it corresponds to the noise of the amplifiers and electronics. The measurements at 4 K and 60 K also include a thermal noise correction (not considered in [20], where the shot noise data were taken at 0.3 K). The shot noise is then normalized by the full shot noise expected for each voltage, calculated from the *I-V* curves for each of the different magnetic states.

Fig. 3(b) compares the bias dependence of the Fano factor in the P state measured at temperatures of 0.3 K, 4 K and 60 K. One can see the presence of strong anomalies in the bias dependence of the Fano factor for biases approximately above 500 mV. The relative magnitude of these quasi-periodic anomalies increases with the applied bias and is weakly dependent on the magnetic



Fig. 4. Comparison of the experimental values (points) and the theoretical results (lines) for the Fano factors measured in the same sample at (a) T = 0.3 K, with fitting parameters g = 100, $\alpha = 0.07$, $\beta_1 = 5.7$, $\beta_2 = 27.8$, and (b) T = 60 K, with g = 100, $\alpha = 0.08$, $\beta_1 = 3.5$, $\beta_2 = 28.3$. The parameters α and $\beta_{1,2}$ describe the barrier asymmetry and the spin filtering of each MgO barrier (see [20] for details).

state. Another interesting feature is the evidence for some low bias suppression of the Fano factor, especially noticeable for the smallest negative biases. We tentatively attribute the above effects to the reduction of the shot noise due to the possible influence of multi-step sequential tunneling through defects. Indeed, numerical calculations predict the presence of FeO and MgO defect states in the Fe/MgO/Fe MTJs at energies below 200 mV [23]. The P state presents the smallest changes in the shot noise at low biases, which could be due to a smaller spin accumulation in this magnetic state.

The quasi-periodic Fano anomalies at highest biases gradually smear out when the samples are heated and become invisible within the error bars when the temperature is around 60 K (Fig. 3(b)). Surprisingly, the QWSs related features in the conductance remain practically unchanged within the same temperature interval (Fig. 3(c)).

VI. THEORETICAL MODEL

The experimental data on the shot n oise vs. temperature have been fitted to the theory presented in [20]. A good agreement has been obtained both for T = 0.3 K and T = 60 K. Fig. 4 presents the fit of the Fano factors estimated for all four magnetic states for (a) T = 0.3 K and (b) T = 60 K. The experimental values of the Fano factor are obtained by averaging over a range of voltages where F is constant. In this case, the chosen range was from +200 mV to +500 mV for both temperatures. The agreement between the experimental data and the theoretical results is quite satisfactory, and it should be emphasized that now the four different magnetic states have been compared to the predicted behavior, whereas in [20] the shot noise was analyzed only for three different magnetic states. As in [20], the relaxation times estimated for this type of asymmetry remain the same (g = 100 for the sample shown in Figs. 2–4).

VII. DISCUSSION

The model described above provides a new method of estimating the spin relaxation time in the central magnetic layer of DMTJs. The theoretical fit of the data obtained for DMTJs with different barrier asymmetries (due to various growth conditions) considered in [22] gives g = 1 - 100 and spin relaxation times τ_S of an order of 10^{-14} – 10^{-12} s, as obtained from the relation $g = d/v_F \tau_S$ assuming the Fermi velocity equal to 10^4 m/s.

It is important, however, to stress the restrictions of the theoretical model used for fitting the experimental data. First, the model does not take into account the bias dependence of the barrier resistance. Apart from this, the shot noise is calculated without taking into account resonant tunneling. The latter, however, should increase when the thickness of the central electrode decreases. As we already mentioned, the model also does not include the influence of resonant tunneling through defect states inside the MgO barriers. Finally, one could also mention the possible spatial fluctuations of such parameters as the barrier resistance (properties of tunneling junctions are mostly determined by small areas of conductive channels). All these factors can be responsible for some deviations of the theoretical curves from the experimental points in Fig. 4.

Finally, the different influence of temperature on the conductivity and shot noise may be accounted for as follows. Resonant tunneling via QWSs is the main factor which affects the conductivity, thus the anomalies in conductance are unchanged up to rather high temperatures, where the peaks in the density of states coming from QWSs decrease only slightly. From the other side, shot noise is influenced mainly by the sequential tunneling mechanism. Seemingly, the sequential tunneling could be affected by the resonant tunneling through QWSs at sufficiently low temperatures, when the spin relaxation length becomes highest.

VIII. CONCLUSION

In conclusion, the shot noise in double-barrier magnetic tunnel junctions could be effectively designed by assuming a proper barrier asymmetry, and also can be controlled by the relative magnetic alignment of the ferromagnetic layers and applied bias. This versatility in control over the most fundamental noise source in electronics could be useful both for vertical (e.g., spin current injection in semiconductors through double MgO barriers) or lateral (e.g., quantum dots) electronic structures.

ACKNOWLEDGMENT

The authors thank D. Herranz and C. C. Bellouard for their help with the experiment. This work was supported by the Spanish MICINN/MINECO (MAT2012-32743, CON-SOLIDER CSD2007-00010, FR2009-0010 grants), Comunidad de Madrid (P2009/MAT-1726), and in part by NCBiR as EU project Era. Net. Rus (SPINBARRIER), and by NCN in Poland as a research project N N202 236540 for the years 2011–2014. C. Tiusan acknowledges the SPINCHAT project ANR-07-BLAN-341 and POS CCE ID. 574, code SMIS-CSNR 12467.

REFERENCES

- W. H. Butler, X.-G. Zhang, T. C. Schulthess, and J. M. MacLaren, "Spin-dependent tunneling conductance of Fe|MgO|Fe sandwiches," *Phys. Rev. B*, vol. 63, p. 054416, 2001.
- [2] J. Mathon and A. Umerski, "Theory of tunneling magnetoresistance of an epitaxial Fe/MgO/Fe(001) junction," *Phys. Rev. B*, vol. 63, p. 220403(R), 2001.
- [3] S. S. Parkin, C. Kaiser, A. Panchula, P. M. Rice, B. Hughes, M. Samant, and S. H. Yang, "Giant tunnelling magnetoresistance at room temperature with MgO (100) tunnel barriers," *Nat. Mater.*, vol. 3, p. 862, 2004.
- [4] S. Yuasa, T. Nagahama, A. Fukushima, Y. Suzuki, and K. Ando, "Giant room-temperature magnetoresistance in single-crystal Fe/MgO/Fe magnetic tunnel junctions," *Nat. Mater.*, vol. 3, p. 868, 2004.
- [5] J. Barnas and A. Fert, "Magnetoresistance oscillations due to charging effects in double ferromagnetic tunnel junctions," *Phys. Rev. Lett.*, vol. 80, p. 1058, 1998.
- [6] T. Nozaki, N. Tezuka, and K. Inomata, "Quantum oscillation of the tunneling conductance in fully epitaxial double barrier magnetic tunnel junctions," *Phys. Rev. Lett.*, vol. 96, p. 027208, 2006.
- [7] Y. Wang, Z.-Y. Lu, X.-G. Zhang, and X. F. Han, "First-principles theory of quantum well resonance in double barrier magnetic tunnel junctions," *Phys. Rev. Lett.*, vol. 97, p. 087210, 2006.
- [8] D. Herranz, F. G. Aliev, C. Tiusan, M. Hehn, V. K. Dugaev, and J. Barnań, "Tunneling in double barrier junctions with "hot spots"," *Phys. Rev. Lett.*, vol. 105, p. 047207, 2010.
- [9] M. Wilczynski, R. Swirkowicz, and J. Barnas, "Phonon-assisted kondo resonance in spin-dependent transport through a quantum dot," *Acta Physica Polonica A*, vol. 115, p. 272, 2009.
- [10] L. Berger, "Multilayer configuration for experiments of spin precession induced by a dc current," J. Appl. Phys., vol. 94, p. 7693, 2003.
- [11] M. Watanabe, J. Okabayashi, H. Toyao, T. Yamaguchi, and J. Yoshino, "Current-driven magnetization reversal at extremely low threshold current density in (Ga, Mn)As-based double-barrier magnetic tunnel junctions," *Appl. Phys. Lett.*, vol. 92, p. 082506, 2008.
- [12] A. Vedyayev, N. Ryzhanova, B. Dieny, and N. Strelkov, "Resonant spin-torque in double barrier magnetic tunnel junctions," *Phys. Lett. A*, vol. 355, p. 243, 2006.
- [13] I. Theodonis, A. Kalitsov, and N. Kioussis, "Enhancing spin-transfer torque through the proximity of quantum well states," *Phys. Rev. B*, vol. 76, p. 224406, 2007.
- [14] Ya. M. Blanter and M. Büttiker, "Enhancing spin-transfer torque through the proximity of quantum well states," *Phys. Rep.*, vol. 336, no. 1, p. 1, 2000.
- [15] B. R. Bulka, J. Martinek, G. Michalek, and J. Barnas, "Shot noise in ferromagnetic single electron tunneling devices," *Phys. Rev. B*, vol. 60, p. 12246, 1999.
- [16] Y. Tserkovnyak and A. Brataas, "Shot noise in ferromagnet—Normal metal systems," *Phys. Rev. B*, vol. 64, p. 214402, 2001.
- [17] G. Iannaccone, G. Lombardi, M. Macucci, and B. Pellegrini, "Enhanced shot noise in resonant tunneling: Theory and experiment," *Phys. Rev. Lett.*, vol. 80, p. 1054, 1998.
- [18] R. Lopez and D. Sanchez, "Nonequilibrium spintronic transport through an artificial kondo impurity: Conductance, magnetoresistance, and shot noise," *Phys. Rev. Lett.*, vol. 90, p. 116602, 2003.
- [19] A. Cottet, W. Belzig, and C. Bruder, "Positive cross correlations in a three-terminal quantum dot with ferromagnetic contacts," *Phys. Rev. Lett.*, vol. 92, p. 206801, 2004.
- [20] J. P. Cascales, D. Herranz, F. G. Aliev, T. Szczepański, V. K. Dugaev, J. Barnań, A. Duluard, M. Hehn, and C. Tiusan, "Controlling shot noise in double-barrier magnetic tunnel junctions," *Phys. Rev. Lett.*, vol. 109, p. 066601, 2012.
- [21] R. Guerrero, F. G. Aliev, Y. Tserkovnyak, T. S. Santos, and J. S. Moodera, "Shot noise in magnetic tunnel junctions: Evidence for sequential tunneling," *Phys. Rev. Lett.*, vol. 97, p. 266602, 2006.
- [22] R. Guerrero, D. Herranz, F. G. Aliev, F. Greullet, C. Tiusan, M. Hehn, and F. Montaigne, "High bias voltage effect on spin-dependent conductivity and shot noise in carbon-doped Fe(001)/MgO(001)/Fe(001) magnetic tunnel junctions," *Appl. Phys. Lett.*, vol. 91, p. 132504, 2007.
- [23] G. X. Du, S. G. Wang, Q. L. Ma, Y. Wang, R. C. C. Ward, X.-G. Zhang, C. Wang, A. Kohn, and X. F. Han, "Spin-dependent tunneling spectroscopy for interface characterization of epitaxial Fe/MgO/Fe magnetic tunnel junctions," *Phys. Rev. B*, vol. 81, p. 064438, 2010.