Shot Noise in Magnetic Tunnel Junctions: Evidence for Sequential Tunneling

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We report the experimental observation of sub-Poissonian shot noise in single magnetic tunnel junctions, indicating the importance of tunneling via impurity levels inside the tunnel barrier. For junctions with weak zero-bias anomaly in conductance, the Fano factor (normalized shot noise) depends on the magnetic configuration being enhanced for antiparallel alignment of the ferromagnetic electrodes. We propose a model of sequential tunneling through nonmagnetic and paramagnetic impurity levels inside the tunnel barrier to qualitatively explain the observations.

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The discovery of the giant magnetoresistance [1] followed by the observation of a large tunneling magnetoresistance (TMR) at room temperature in magnetic tunnel junctions (MTJ's) [2] have boosted interest in spin-dependent electron transport in magnetic nanostructures, especially in the spin-dependent tunneling phenomena [3,4]. During recent years, there has been a growing interest in controlling the TMR and also the statistics of tunneling events in MTJ's by nanostructuring of the insulating barrier [5]. A variety of new electron-correlation mechanisms have been proposed, typically based on transport through double MTJ's with either an open or Coulomb-blockaded quantum dot (QD) contacted by ferromagnetic electrodes. Electric shot noise (SN) is a powerful tool for studying correlations of tunneling processes in nanostructures beyond the capabilities of dc measurements [6]. The growing list of theoretically investigated topics regarding spin-dependent shot noise includes the noise asymmetry between parallel (P) and antiparallel (AP) ferromagnetic (FM) alignment [7] as well as continuous variation of the SN over the relative angle between FM electrodes [8], SN through an artificial (QD) Kondo impurity [9] contacted by magnetic leads, shot-noise enhancement by dynamic spin blockade in tunneling through a small QD [10], and shot noise for spin-polarized and entangled electrons with spin-orbit interaction in the leads [11]. The scope of experimental efforts [12–14] has however so far been much more limited and inconclusive with regard to the nature of tunneling electron correlations even in the conceptually simplest spintronic devices, viz., MTJ's, as manifested by shot-noise measurements.

Current fluctuations due to discreteness of electron charge flowing through the structure out of equilibrium, which provide the shot noise, contain information not accessible by time-independent conductance. Sensitivity to quantum statistics, interference, and interactions between electrons passing through the device has made SN an effective tool for investigating quantum transport in meso- and nanostructures [6]. In the absence of any correlations, Poissonian shot noise is practically frequency independent at low frequencies with the noise power given by $S = 2eI$, in terms of the average current I. The Fano factor $F = S/2eI$ representing normalized shot noise is in general lowered below 1 for noninteracting electrons due to fermionic statistics. Electron-electron interactions can either further suppress or enhance the Fano factor (even beyond the Poissonian value).

Despite the theoretical excitement about perspectives of using the shot noise for investigation of spin-polarized electrons, behavior of the SN even in simple nonstructured MTJ's remains unclear. Jiang *et al.* [12] reported an observation of the "full" SN (i.e., $F \approx 1$) in MTJ's with AP alignment of electrodes. Later, the same group [13] measured a strong suppression (down to $F \approx 0.45$) of the SN in magnetic tunnel junctions, which was not understood. Our Letter reports the first systematic investigation of the tunneling statistics in a magnetic tunneling device by measuring shot noise in Co(80 Å)|Al_2O_3(14 Å)|Py(100 Å) MTJ's with and without Cr doping of the insulating barrier. We demonstrate a decrease of the Fano factor and its dependence on the alignment of the ferromagnetic electrodes for certain barrier conditions.

Details of sample preparation have been published previously [15]. For Cr-doped samples, the tunnel barriers were deposited in two steps. After deposition of the underlying Co electrode, a first tunnel barrier was formed by deposition and subsequent oxidation of 7–9 Å of Al. Subsequently, submonolayer amounts of Cr were deposited on the Al_2O_3 surface, followed by a second Al layer deposition (5–7 Å) and oxidation, resulting in a "δ-doped" Al_2O_3|Cr|Al_2O_3 tunnel barrier. The noise measurements use a setup described in Ref. [16], which employs the cross-correlation method. This technique removes uncorrelated noise from the amplifiers and the noise of the leads and takes into account nonlinearity of the dynamic resistance while converting the obtained voltage noise into current noise. Out of 13 samples investigated, the shot noise was measured for 11 MTJ's: 5 without and 6 with δ-layer of Cr in the middle of the barrier, ranging between 0.2 and 1.2 Å in thickness.

Figure 1 shows typical electron transport characteristics of the studied MTJ's. The dynamic tunneling resistance vs bias V [Figs. 1(a) and 1(b)] measured at three temperatures for P alignment proves pinhole-free MTJ's [3]. For all MTJ's studied, an asymmetric parabolic conductance background [17] plus a zero-bias anomaly (ZBA) below $T \sim 77$ K, appeared in the resistance of the junction (R_J) [Figs. 1(a) and 1(b)]. Presently, there exist several possible explanations of the ZBA's in MTJ's [18,19], which consider magnon- or phonon-assisted tunneling or two-step tunneling through impurities inside the tunnel barrier which are also coupled to some additional degrees of freedom. Simultaneous ZBA and SN measurements on our samples suggest the ZBA is provided by sequential tunneling through impurities accompanied with spin flips.

Doping of the barrier with Cr enhances the normalized ZBA, although this trend presents rather large dispersion [Fig. 1(d)]. Conductivity and TMR are generally suppressed when Cr thickness is increased, but the relations between these parameters and the nominal Cr concentration are not strictly monotonic. We have found, however, that the changes in the TMR are correlated with those of the tunneling resistance (see below). This can be understood as follows: As the barrier width and the resistance increase, the relative role of two-step tunneling increases, which generally reduces the TMR. The TMR is also monotonically reduced with the applied voltage both for the Cr-free and Cr-doped MTJ's [see Fig. 1(c)], in accordance with the previous reports [18].

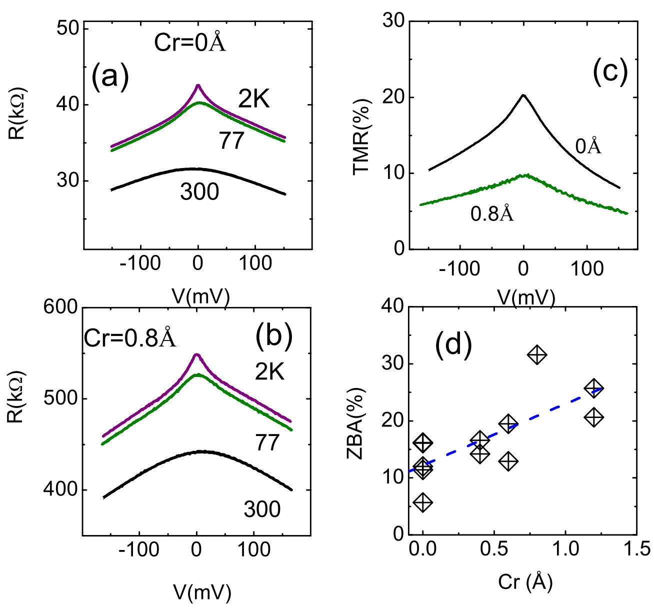

FIG. 1 (color online). Typical dynamic resistance obtained in the P state for the Cr-free (a) and Cr-doped (b) junctions with 0.8 Å δ-layer measured at 300, 77, and 2 K. (c) Reduction of the TMR with applied voltage for Cr-free and Cr-doped MTJ's at $T = 2$ K. (d) Dependence of the ZBA(%) $= 100[R(0\text{ mV}) - R(100\text{ mV})]/R(100\text{ mV})$ determined for the P alignment on Cr (at 2 K).

The measured low-frequency noise has a typical form for MTJ's, with the $1/f$ noise dominating at $f < 100$ Hz and the "white" noise dominating at $f \gtrsim 100$ Hz. Figure 2(a) shows a typical voltage noise for the frequency and bias range where the $1/f$ noise does not affect the data and the applied bias ($eV \gg k_B T$) ensures that SN presents the dominant contribution to the total noise. Figure 2(b) shows a typical dependence of F on bias. For most of the undoped MTJ's, the Fano factor was reduced below the Poissonian value ($F < 1$), while for the Cr-doped MTJ's F was always close to 1.

Figure 3(a) shows the TMR and the Fano factor for the P alignment as a function of the resistance by area product ($R \times A$) at $T = 2$ K. The Fano factor was averaged over the range 40–120 mV where it is nearly bias independent. For the undoped MTJ's in the range where TMR is only weakly reduced with the product $R \times A$ ($<10^4$ M$\Omega\,\mu$m^2), we observed a gradual suppression of the Fano factor down to $F \sim 0.65$. Doping of the barrier with Cr further increases the tunneling resistance and restores the Poissonian SN ($F \sim 1$). The suppression of F in a certain tunneling resistance range is not accompanied by the appearance of random telegraph noise as in Ref. [14], reduced TMR [20], or by metallic temperature dependence $R(T)$, clearly ruling out pinholes or hot spots across the barrier. Figure 3(b) shows the normalized AP-P F asymmetry as a function of the normalized ZBA for the P alignment. Surprisingly, we find that F depends on the alignment of the electrodes with $F_{AP}/F_P > 1$ only in the MTJ's with a weak zero-bias anomaly and becomes nearly independent of the alignment above some threshold value of the ZBA. We stress that the observed Fano factor asymmetry reflects only alignment of the FM electrodes, but not orientation of the magnetic field.

Previous studies of the shot noise in nonmagnetic TJ's with Al_2O_3 barrier have observed Poissonian value $F \simeq 1$ [21]. It was reported, however, that for nonmagnetic TJ's

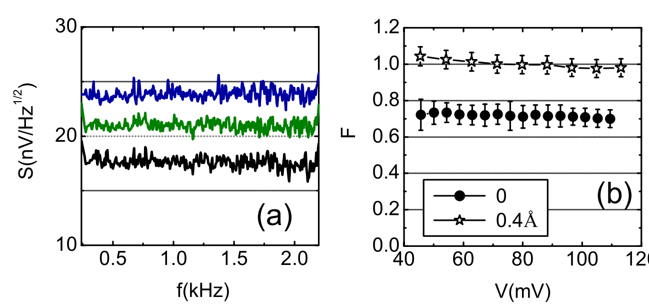

FIG. 2 (color online). (a) Typical voltage noise measured for a Cr-free MTJ at $T = 2$ K with the applied currents (from bottom to up) of 3.4, 5 and 6 μA. (b) Voltage dependence of F on bias, for the Cr-free (filled) and 0.4 Å Cr-doped (open stars), also measured at $T = 2$ K. The error bars show standard deviations.

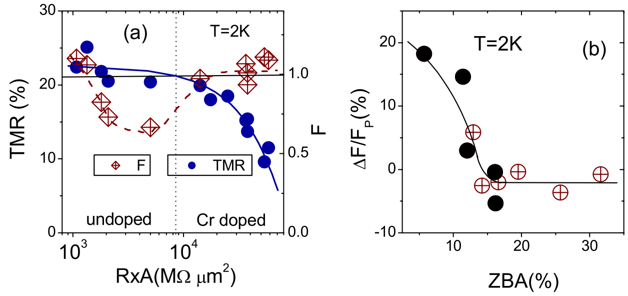

FIG. 3 (color online). (a) Dependence of the TMR and F (for the P state) on the resistance area product ($R \times A$). Solid horizontal line marks $F = 1$. Vertical dashed line separates the Cr-free and Cr-doped regions. (b) Dependence of the relative variation of F with alignment $\Delta F/F_{\mathrm{P}}(\%) = 100(F_{\mathrm{AP}} - F_{\mathrm{P}})/F_{\mathrm{P}}$ on the relative strength of the ZBA. Solid symbols point to the undoped samples. The lines are guides for the eye.

with SiO_2 barrier, tunneling through localized states within the barrier, could indeed account for the measured reduced F [22]. In the following, we consider two simple models for sequential tunneling via an island inside the tunnel barrier (see Fig. 4), which capture some qualitative aspects of our measurements. First, consider tunneling through a normal region (R_M) inside the tunnel barrier [Fig. 4(a)]. Neglecting charging effects, we can simply sum the contributions to the (averaged) current and noise for the two spin species. To this end, suppose R_M is coupled asymmetrically to the left and right reservoirs (R_L and R_R) with the respective spin-dependent conductances given by

$$g_{L\uparrow} = g/\sqrt{\beta} \quad \text{and} \quad g_{R\uparrow} = g\sqrt{\beta},$$
$$g_{L\downarrow} = \alpha g/\sqrt{\beta} \quad \text{and} \quad g_{R\downarrow} = \alpha g\sqrt{\beta}. \tag{1}$$

β is a dimensionless left-right asymmetry parameter and α characterizes spin polarization. The charge current at the voltage bias V is given by $I = igV$, parametrized by a dimensionless current i that depends on α and β only. Let us

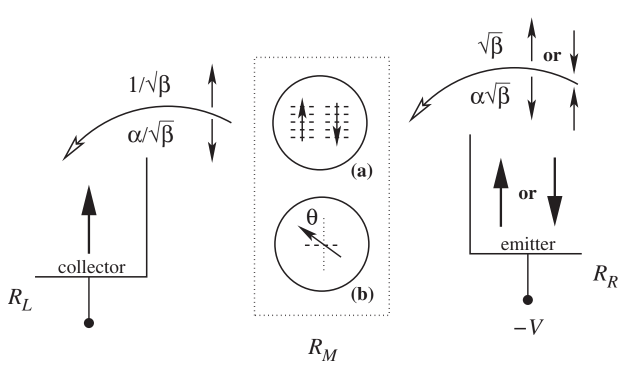

FIG. 4. Two models: (a) a large normal region R_M such that electrons can be treated as noninteracting, coupled to biased emitter and collector reservoirs in P or AP configuration, (b) R_M is a single spin-polarized impurity level that can hold only one electron with spin at a (random) angle θ with respect to the collector magnetization.

furthermore write the zero-frequency shot noise as $S = 2esgV$. The Fano factor thus becomes $F = S/(2eI) = s/i$.

Let us recall first that, in general, noninteracting spinless electrons in double-barrier structures have the series conductance and F (for spinless electrons)

$$G = g_L g_R/(g_L + g_R), \quad F = (g_L^2 + g_R^2)/(g_L + g_R)^2, \tag{2}$$

which are valid not only for large semiclassical R_M but also for sequential tunneling through a small R_M described by master-equation approach, in which case g's become respective transition rates instead of the tunnel-barrier conductances [6]. Summing corresponding current and noise for the two spin channels in the P configuration (neglecting correlations between two spin species), one trivially obtains for the current and F [6]

$$i_{\mathrm{P}} = (1+\alpha)\sqrt{\beta}/(1+\beta), \quad F_{\mathrm{P}} = (1+\beta^2)/(1+\beta)^2. \tag{3}$$

In the AP configuration,

$$i_{\mathrm{AP}} = \alpha(1+\alpha)(1+\beta)\sqrt{\beta}/[(\alpha+\beta)(1+\alpha\beta)],$$
$$F_{\mathrm{AP}} = \frac{\alpha^2(1+2\beta-2\beta^2+2\beta^3+\beta^4)}{(\alpha+\beta)^2(1+\alpha\beta)^2} + (\alpha \leftrightarrow \beta), \tag{4}$$

where $(\alpha \leftrightarrow \beta)$ is the same as the first summand but with α and β interchanged. These results are plotted in Fig. 5(a) for $\alpha = 1/5$. Note that $F_{\mathrm{AP}} - F_{\mathrm{P}} > 0$ for $\beta \sim 1$, roughly corresponding to the center of the junction for the R_M location, which is the region contributing the largest current, see Fig. 5(a). The Fano factor asymmetry is reversed closer to the junction interfaces where the tunneling is asymmetric.

Consider now hopping through a single level that can hold only one extra electron, see Fig. 4(b). If there is a large exchange-energy splitting along certain direction θ, one could imagine a situation when only spins polarized along θ are energetically allowed to tunnel through. We can then

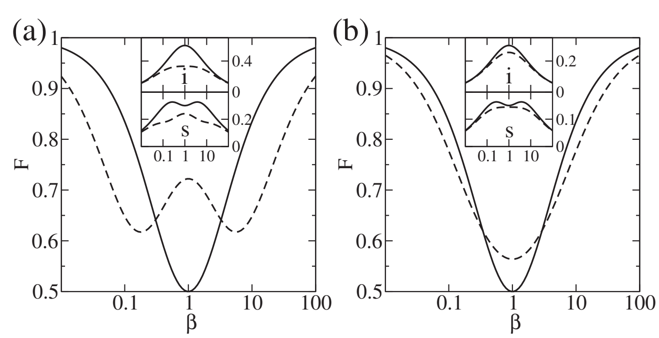

FIG. 5. Fano factor as a function of the left-right asymmetry parameter β setting $\alpha = 1/5$ for the two models sketched in Fig. 4. Solid lines are for the P and dashed lines for the AP magnetic configurations. Insets show the dimensionless current i and noise $s = Fi$ defined in the text. Note the logarithmic scale for β: Assuming tunnel rates depend exponentially on the barrier thickness, this corresponds to a linear scale for the R_M position inside the tunnel junction (small β corresponding to the proximity to R_L and large β to R_R).

calculate the current and noise using Eqs. (2) where the rates g_L and g_R now depend on θ and the relative magnetic orientation in the leads [23]:

$$g_L = g_{L\uparrow}(1 + \cos\theta)/2 + g_{L\downarrow}(1 - \cos\theta)/2,$$
$$g_R = g_{R\uparrow}(1 \pm \cos\theta)/2 + g_{R\downarrow}(1 \mp \cos\theta)/2 \quad (5)$$

for the P (AP) configuration. Assuming θ is random, we average the current and noise: $\langle \ldots \rangle_\theta = (1/2) \times \int_{-1}^{1} d(\cos\theta)\ldots$. This results in simple expressions for the P MTJ:

$$i_P = (1 + \alpha)\sqrt{\beta}/[2(1 + \beta)],$$
$$F_P = (1 + \beta^2)/(1 + \beta)^2, \quad (6)$$

which are the same as just averaging over $\theta = 0$ and π. There is no simple analytic form for the current and noise in the AP case. We plot the results in Fig. 5(b). Notice that the AP-P asymmetry is significantly reduced in comparison to Fig. 5(a).

In undoped MTJ's, we measured typically $F_{AP} > F_P$ and both are significantly suppressed below 1, apart from the thinnest tunnel barrier. Both of these findings are consistent with the results in Fig. 5 for tunneling predominantly through impurities in the middle of the barrier. Since F for tunneling through uniformly distributed pointlike localized states is in general $3/4$ (in the absence of hopping correlations between the two spin species) [24], which, in particular, applies to both models in Fig. 4, the AP-P asymmetry would require some structural preference towards tunneling through the middle of the barrier. The Fano factor is reduced to $F \sim 3/4$, as shown in Fig. 3(a), as the tunnel barrier becomes wider and the role of the two-step tunneling processes become relatively more important. The tunneling resistance does not indicate variable-range hopping involving multistep tunneling, which was observed for wider tunnel barriers [25]. Observation of Poissonian noise after Cr doping could be due to an offset in Cr deposited nominally in the center of the junction, which leads to systematically asymmetric hopping. Finally, the observed correlation in the AP-P Fano factor asymmetry and the ZBA [Fig. 3(b)] suggest that an inelastic spin-flip mechanism in the barrier may be responsible for concurrent reduction of the former and enhancement of the latter.

In summary, first systematic shot-noise measurements in a magnetic tunnel junction show an evidence for sequential tunneling mediated by defects. We demonstrate for the first time that electron tunneling statistics can be manipulated by an applied magnetic field due to their dependence on the relative orientation of ferromagnetic electrodes and also by deliberately doping the tunnel barrier with impurities. Control over the sequential tunneling could find applications in optimizing signal-to-noise ratio in magnetoelectronic devices and provide a new tool for investigating spin-dependent transport of electrons injected by ferromagnetic electrodes.

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[1] M. N. Baibich *et al.*, Phys. Rev. Lett. **61**, 2472 (1988).
[2] J. S. Moodera *et al.*, Phys. Rev. Lett. **74**, 3273 (1995); T. Miyazaki and N. Tezuka, J. Magn. Magn. Mater. **139**, L231 (1995).
[3] J. S. Moodera, J. Nassar, and G. Mathon, Annu. Rev. Mater. Sci. **29**, 381 (1999).
[4] E. Y. Tsymbal, O. N. Mryasov, and P. R. LeClair, J. Phys. Condens. Matter **15**, R109 (2003); S. Yuasa *et al.*, Nat. Mater. **3**, 868 (2004); S. S. P. Parkin *et al.*, *ibid.* **3**, 862 (2004).
[5] J. Barnaś and A. Fert, Phys. Rev. Lett. **80**, 1058 (1998); S. Takahashi and S. Maekawa, *ibid.* **80**, 1758 (1998).
[6] Ya. M. Blanter and M. Büttiker, Phys. Rep. **336**, 1 (2000).
[7] B. R. Bulka *et al.*, Phys. Rev. B **60**, 12 246 (1999).
[8] Y. Tserkovnyak and A. Brataas, Phys. Rev. B **64**, 214402 (2001).
[9] R. Lopez and D. Sanchez, Phys. Rev. Lett. **90**, 116602 (2003).
[10] A. Cottet, W. Belzig, and C. Bruder, Phys. Rev. Lett. **92**, 206801 (2004).
[11] J. C. Egues, G. Burkard, and D. Loss, Phys. Rev. Lett. **89**, 176401 (2002).
[12] L. Jiang *et al.*, Phys. Rev. B **69**, 054407 (2004).
[13] L. Jiang *et al.*, Proc. SPIE-Int. Soc. Opt. Eng. **5469**, 13 (2004).
[14] E. R. Nowak, M. B. Weissman, and S. S. P. Parkin, Appl. Phys. Lett. **74**, 600 (1999).
[15] R. Jansen and J. S. Moodera, Phys. Rev. B **61**, 9047 (2000).
[16] R. Guerrero *et al.*, Appl. Phys. Lett. **87**, 042501 (2005).
[17] W. F. Brinkman, R. C. Dynes, and J. M. Rowell, J. Appl. Phys. **41**, 1915 (1970).
[18] S. Zhang *et al.*, Phys. Rev. Lett. **79**, 3744 (1997); J. S. Moodera *et al.*, *ibid.* **80**, 2941 (1998).
[19] J. Zhang and R. White, J. Appl. Phys. **83**, 6512 (1998); L. Sheng, D. Y. Xing, and D. N. Sheng, Phys. Rev. B **70**, 094416 (2004).
[20] P. K. George *et al.*, Appl. Phys. Lett. **80**, 682 (2002).
[21] L. Spietz *et al.*, Science **300**, 1929 (2003).
[22] G. Iannaccone *et al.*, IEEE Trans. Electron Devices **50**, 1363 (2003).
[23] J. C. Slonczewski, Phys. Rev. B **71**, 024411 (2005).
[24] Y. V. Nazarov and J. J. R. Struben, Phys. Rev. B **53**, 15 466 (1996).
[25] Y. Xu, D. Ephron, and M. R. Beasley, Phys. Rev. B **52**, 2843 (1995).