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Nonmonotonic angular magnetoresistance in asymmetric spin valves

Jan Manschot,^{1,2} Arne Brataas,² and Gerrit E. W. Bauer¹

¹*Department of Nanoscience, Delft University of Technology, 2628 CJ Delft, The Netherlands*

²*Department of Physics, Norwegian University of Science and Technology, N-7491 Trondheim, Norway*

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The electric resistance of ferromagnet/normal-metal/ferromagnet perpendicular spin valves depends on the relative angle between the magnetization directions. In contrast to common wisdom, this angular magnetoresistance is found to be not necessarily a monotone function of the angle. The parameter dependence of the global resistance minimum at finite angles is studied and the conditions for experimental observation are specified.

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The discovery of the giant magnetoresistance (GMR) (Ref. 1) has driven much of the current research to enrich the functionalities of electronic circuits and devices employing ferromagnetic elements. The current perpendicular to plane (CPP) transport technique²⁻⁴ turned out to be especially suited to study the physics of spin dependent transport. Nanostructured and perpendicular spin valves are ideal devices to investigate the current-induced magnetization reversal,⁵ which has potential applications for magnetic random access memories. These structures allow the measurement of the angular magnetoresistance (AMR) (Refs. 6,7) introducing an analog degree of freedom between the conventional parallel versus antiparallel digital configurations. A semiclassical theoretical treatment of the AMR leads to the concept of a spin-mixing conductance⁸ that turned out useful for phenomena like the spin torque^{9,10} and interface-enhanced Gilbert damping.¹¹

This Brief Report addresses the AMR of asymmetric perpendicular spin valves. We show that the parallel magnetization configuration of ferromagnet (F)/normal-metal (N)/ferromagnet heterostructures does not necessarily correspond to the minimal resistance. This nonmonotonic behavior requires a redefinition of the GMR ratio in terms of the global maximum and minimum resistances instead of those for parallel and antiparallel configurations. We discuss how to optimize the conditions for an experimental observation and demonstrate that the spin torque is strongly affected by the asymmetry as well.

First, we summarize necessary concepts from Ref. 12 for resistive elements such as an interface between a monodomain ferromagnet with magnetization parallel to the unit vector $\mathbf{m}$ and a normal metal. The charge and spin current, I_c and I_s , driven by a potential and spin accumulation bias, $\Delta\mu_c$ and $\Delta\mu_s$, read

$$I_c = \frac{e}{h} [(g^{\uparrow\uparrow} + g^{\downarrow\downarrow}) \Delta\mu_c + (g^{\uparrow\uparrow} - g^{\downarrow\downarrow}) \mathbf{m} \cdot \Delta\boldsymbol{\mu}_s], \quad (1)$$

$$I_s = \frac{1}{4\pi} \mathbf{m} [(g^{\uparrow\uparrow} - g^{\downarrow\downarrow}) \Delta\mu_c + (g^{\uparrow\uparrow} + g^{\downarrow\downarrow}) \mathbf{m} \cdot \Delta\boldsymbol{\mu}_s] + \frac{1}{4\pi} 2\text{Re}(g^{\uparrow\downarrow}) \mathbf{m} \times (\Delta\boldsymbol{\mu}_s \times \mathbf{m}), \quad (2)$$

where $g^{\uparrow\uparrow}$ and $g^{\downarrow\downarrow}$ are the conductances for electrons with majority and minority spin, respectively, and $g^{\uparrow\downarrow}$ is the mixing conductance for a spin current polarized transverse to the magnetization. We disregarded $\text{Im}(g^{\uparrow\downarrow})$, which for metallic interfaces is usually smaller than 10% of $\text{Re}(g^{\uparrow\downarrow})$.^{9,13} It is convenient to introduce $g = g^{\uparrow\uparrow} + g^{\downarrow\downarrow}$, $p = (g^{\uparrow\uparrow} - g^{\downarrow\downarrow})/g$, and $\eta = 2g^{\uparrow\downarrow}/g$, where g is the total conductance, p the polarization, and η the relative mixing conductance.

Let us examine a two terminal system (F-N-F) as shown in Fig. 1. The contacts need not be identical; the conduction parameters are summarized as G_L and G_R . The electric resistance as function of the angle between the magnetization directions of the reservoirs can simply be calculated using Eqs. (1) and (2), assuming charge and spin conservation on the normal-metal node. For a *symmetric* structure ($G_L = G_R = G$) the resistance $R(\theta)$ reads¹²

$$\frac{e^2}{h} R(\theta) = \frac{2}{g} \frac{\tan^2 \theta/2 + \eta}{(1 - p^2) \tan^2 \theta/2 + \eta}. \quad (3)$$

If necessary, spin-flip processes in the normal metal can be included.¹² A finite angle between the magnetizations causes a spin accumulation on the normal-metal node. Since we disregard the imaginary part of the mixing conductance, it lies in the plane of the magnetization vectors. The resistance increases with increasing spin accumulation, whose creation costs energy, and thus with θ . Therefore the resistance is minimal when the magnetizations are parallel and maximal for $\theta = \pi$. The mixing conductance can be interpreted as an additional channel for dissipating the spin accumulation on the normal-metal node for $0 < \theta < \pi$; an increasing mixing

FIG. 1. Schematic picture of a perpendicular spin valve biased by a voltage difference V . θ is the angle between the magnetization directions of both reservoirs. The reservoirs and contacts need not be identical; the conduction parameters are summarized as G_L and G_R .

FIG. 2. Schematic picture of a thin-film pillar with two ferromagnetic (F), three normal-metal (N) layers, and two normal-metal reservoirs.

conductance will therefore reduce the total resistance. This is the mechanism behind deviations of the AMR from a simple $\cos^2\theta/2$ behavior, which can be used to determine the mixing conductance from experimental curves.^{7,10}

In the following, we focus on an asymmetric configuration with $G_L \neq G_R$. The asymmetry in conductance ($g_L \neq g_R$) causes a charge accumulation on the normal-metal node. Similarly, when $p_L \neq p_R$, a spin accumulation is excited on the normal-metal node even for $\theta = 0$. We find here that configurations with $\theta \neq 0$ may correspond to a spin accumulation that is smaller than that of the parallel one, and therefore a global resistance minimum at finite angles. The recipe for a significant effect is a large polarization of the current by the source contact (e.g., $p_L \approx 1$) and efficient dissipation of the spin accumulation for a finite angle θ by a large mixing conductance ($\eta_R > 1$) of the drain. The polarization direction of the spin current differs from the magnetization directions for finite angles θ as in the symmetric case, but the noted asymmetry forces it to be close to the magnetization direction of the source contact. A large mixing conductance $g^{\uparrow\downarrow}$ favors the transverse over the longitudinal spin current. Spins on the normal-metal node therefore escape easily and the reduced spin accumulation is equivalent to a decrease of the total resistance. This interplay between spin accumulation and magnetization angles strongly modifies the total AMR profile.

Unfortunately, the exact equations for $R(\theta)$ are not very transparent. A perturbation approach to these equations is not helpful because no small parameters can be identified for the experimentally relevant metallic structures. However, we did find relatively simple analytical expressions for the angle θ_m of the global resistance minimum by solving $dR(\theta_m)/d\theta_m = 0$. Another simple expression is obtained for the maximal AMR in the limit $\eta_R \gg 1$.

We derive that, besides $\theta = 0, \pi$, the resistance may have extrema at two additional angles

$$\cos \theta_{m1} = \left(\frac{p_R}{p_L} \right) \left(\frac{1 + \left(\frac{g_L}{g_R} \right) \frac{1 - p_L^2}{\eta_R}}{1 - \frac{1 - p_R^2}{\eta_R}} \right), \quad (4)$$

$$\cos \theta_{m2} = \left(\frac{p_L}{p_R} \right) \left(\frac{1 + \left(\frac{g_R}{g_L} \right) \frac{1 - p_R^2}{\eta_L}}{1 - \frac{1 - p_L^2}{\eta_L}} \right), \quad (5)$$

where the absolute value of $\cos \theta_m$ must be smaller than unity, which is clearly not the case for a symmetrical spin valve. The condition for one extra extremum is easily fulfilled.

Two additional extrema are not consistent with the condition $\eta_L, \eta_R > 1$, which rigorously holds for high contact resistances,¹² but not necessarily for highly transparent interfaces.¹⁰ It can be proved that when one extremum exists and $\eta_L, \eta_R > 1$, the extremum is the global minimum and located in the interval $0 < \theta < \pi/2$. When $\eta_R < 1 - p_R^2$ (that does not seem very likely for metals), an additional extremum may exist in the interval $\pi/2 < \theta < \pi$. It turns out to be a maximum that can be understood in the same way as the minimum. Additional minima and maxima may even coexist for specific parameter combinations, which do not appear relevant for metallic spin valves, however.

The position of the global minimum does not depend on η of the source contact. The source polarizes the current through the total structure parallel to its magnetization, therefore the source η does not play a role at all. The component of the spin current orthogonal to the magnetization is called the spin torque⁹ acting on this magnetization, since it is absorbed by the magnetic order parameter and may excite the magnetization when exceeding a threshold value.^{5,14,15} In the global minimum the spin torque on the source magnetization vanishes with the transverse component of the spin current. The spin torque on the drain is large, but not at the maximum as a function of θ .

Let us choose the left lead to be the polarizing source ($p_L > p_R$) and the right lead to be the dissipating drain ($\eta_L > \eta_R$). The condition for a noncollinear resistance minimum is now

$$\left| \left(\frac{p_R}{p_L} \right) \left(\frac{1 + \left(\frac{g_L}{g_R} \right) \frac{1 - p_L^2}{\eta_R}}{1 - \frac{1 - p_R^2}{\eta_R}} \right) \right| < 1. \quad (6)$$

When the second factor is larger than 1 (true for $\eta_R > 1$), only a polarization ratio $p_L/p_R > 1$ can save this inequality. The condition is never fulfilled when the left-hand side of Eq. (6) diverges:

$$\frac{1 - p_R^2}{\eta_R} = \frac{2 \left(\frac{1}{g_R^{\uparrow\uparrow}} + \frac{1}{g_R^{\downarrow\downarrow}} \right)^{-1}}{g_R^{\uparrow\downarrow}} \approx 1. \quad (7)$$

Therefore $g_R^{\uparrow\downarrow}$ should be considerably larger than $g_R^{\uparrow\uparrow}$ and $g_R^{\downarrow\downarrow}$. When the average conductance of the source is smaller than the mixing conductance of the drain, the numerator

FIG. 3. Angular dependence of the thin-film pillar resistance. θ is the angle between the magnetization directions of both layers.

$$\left(\frac{g_L}{g_R}\right) \frac{1-p_L^2}{\eta_R} = \frac{2\left(\frac{1}{g_L^{\uparrow\uparrow}} + \frac{1}{g_L^{\downarrow\downarrow}}\right)^{-1}}{g_R^{\uparrow\downarrow}}, \quad (8)$$

reduces $\cos \theta_m$, and hence increases θ_m .

The GMR ratio is usually defined in terms of the resistance in parallel or antiparallel configurations, in terms of the previously introduced parameters,

$$\text{GMR}^* = \frac{R^{\text{ap}} - R^{\text{p}}}{R^{\text{ap}}} = \frac{g_L g_R [(p_L + p_R)^2 - (p_L - p_R)^2]}{(g_L + g_R)^2 - (g_L p_L - g_R p_R)^2}, \quad (9)$$

where R^{ap} and R^{p} are the resistances for antiparallel and parallel configurations, respectively. Hence, the GMR ratio increases when the total polarization increases, as expected. However, when the difference between the polarizations of both sides is large, GMR^* decreases because of a larger spin accumulation on the normal-metal node for $\theta=0$, as noted above. Since now $R^{\text{ap}} - R^{\text{p}}$ is no longer the maximal resistance difference, a new definition for the magnetoresistance is appropriate in terms of the global maximum (for which we still take the antiparallel configuration) and the newly found global minimum. In the limit of large $\eta_R \gg 1$ we arrive at the simple result

$$\text{GMR} = \frac{R^{\text{ap}} - R^{\text{m}}}{R^{\text{ap}}} = \text{GMR}^* \frac{(p_L + p_R)^2}{4p_L p_R}, \quad (10)$$

where R^{m} is the global minimum of the resistance. It can easily be verified that GMR is indeed larger than GMR^* .

Next we investigate the conditions under which this enhanced magnetoresistance can be measured in magnetic spin valves with a current perpendicular to plane geometry.² Even for identical magnetic layers an asymmetry can be realized by a spin independent resistance or tunnel barrier at the outside of one of the magnetic films, as long as the spin-diffusion length is larger than the total bilayer. Such an additional series resistor then effectively decreases p , $g^{\uparrow\uparrow}$, and $g^{\downarrow\downarrow}$ of this magnet. Because the spin current normal to the

FIG. 4. Normalized spin torque on the left magnetization as a function of θ .

magnetization is absorbed by the magnet⁵ over a couple of monolayers, $g^{\uparrow\downarrow}$ is not modified by the extra resistance.⁹ $g^{\uparrow\uparrow}$ thus can indeed be engineered to be larger than $g^{\uparrow\downarrow}$ for a given contact such that a nonmonotonic AMR can be expected.

Spin dependent bulk resistances contribute to the AMR over thicknesses smaller than the spin-diffusion length. Copper and cobalt have relatively large spin-flip lengths, 250 nm and 50 nm, respectively, which makes them useful materials to explore this effect. Al_2O_3 tunnel barriers are routinely used for tunnel MR studies and suitable materials for the present purposes.

The full AMR profile can best be calculated numerically. We consider a structure consisting of two identical cobalt layers (thickness is 3 nm) separated by a thin copper layer (10 nm). Both magnets are sandwiched by tunnel junctions, another copper layer, and finally normal-metal reservoirs as sketched in Fig. 2. Bulk resistances of copper and cobalt are disregarded because they are relatively small. G_F symbolizes all conductance parameters of a copper-cobalt interface, G_L and G_R stand for the outer normal resistances including the tunnel junctions. For interfaces between a ferromagnet and a normal metal, $g^{\uparrow\downarrow}$ ($\sim$ number of modes in the normal metal) usually lies between $g^{\uparrow\uparrow}$ and $g^{\downarrow\downarrow}$. For a Co/Cu interface g is typically 1413×10^3 (for an interface cross section of $140 \times 90 \text{ nm}^2$), $p=0.75$ (Ref. 4) and $\eta=0.38$ (Ref. 9); these values include the Boltzmann corrections for transparent interfaces.¹⁰ In order to compare configurations with different values for G_L and G_R , its series resistance is assumed constant at $1/G_L + 1/G_R = 0.37 \Omega$.

The computed AMR is presented in Fig. 3 for three different ratios $G_L : G_R$. When G_L differs sufficiently from G_R , the global minimum shifts away from the parallel configuration, as predicted. The position of the global minimum θ_m increases with increasing polarization contrast. The GMR ratio increases as well, which is in qualitative agreement with Eq. (10).

Finally we compute the spin torque, i.e., the transverse component of the spin current. The spin torque in spin valves is governed by a similar expression as the charge current¹⁰

and is strongly affected by the asymmetry as well. It is convenient to normalize the spin torque by the charge current:

$$i_s = \frac{|\mathbf{m} \times (\mathbf{I}_s \times \mathbf{m})|}{|I_c|}. \quad (11)$$

In Fig. 4, i_s of the left magnetization is plotted as a function of θ and different G_L/G_R ratios. The zero points in the interval $0 < \theta < \pi/2$ illustrate that when the left side is the polarizing source, the spin torque at the global minimum vanishes, which agrees with the finding that the resistance minimum is not a function of η_L . A zero point of the spin torque in the interval $0 < \theta < \pi/2$ is previously reported by Kovalev *et al.*;¹⁶ their observation can be attributed as well to asymmetry in the multilayer, although asymmetry is not explicitly mentioned in Ref. 16. We observe in Fig. 4 that the spin torque is strongly enhanced when $G_L/G_R \rightarrow 0$ because the relative mixing conductance of the left-hand side is then highly increased, which physically means that the spin accumulation on the normal-metal node easily can be dissipated.

To summarize, we have shown that the AMR of perpendicular spin valves can be a nonmonotonic function when the contacts between the central normal-metal node and the outer ferromagnets differ. An analytical expression is derived for the angle θ_m at which the magnetoresistance has its global minimum. A definition for the GMR ratio is proposed to take this effect into account. This GMR ratio is now larger than the conventional definition in terms of the resistance of parallel and antiparallel configurations. The spin torque in asymmetric structures is also importantly modified.

Note added. After submission of this paper, A. Shpiro kindly pointed out to us that in the method of Ref. 17, valid in the limit of small exchange splittings, a nonmonotonic AMR can be obtained as well.

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