Artificial ferroelectricity due to anomalous Hall effect in magnetic tunnel junctions

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We theoretically investigated Anomalous Hall Effect (AHE) and Spin Hall Effect (SHE) transversally to the insulating spacer O, in magnetic tunnel junctions of the form F/O/F where F are ferromagnetic layers and O represents a tunnel barrier. We considered the case of purely ballistic (quantum mechanical) transport, taking into account the asymmetric scattering due to spin-orbit interaction in the tunnel barrier. AHE and SHE in the considered case have a surface nature due to proximity effect. Their amplitude is in first order of the scattering potential. This contrasts with ferromagnetic metals wherein these effect are in second (side-jump scattering) and third (skew scattering) order on these potentials. The value of AHE voltage in insulating spacer may be much larger than in metallic ferromagnetic electrodes. For the antiparallel orientation of the magnetizations in the two F-electrodes, a spontaneous Hall voltage exists even at zero applied voltage. Therefore an insulating spacer sandwiched between two ferromagnetic layers can be considered as exhibiting a spontaneous ferroelectricity.

Keywords: Hall effect, anomalous Hall effect, spin Hall effect, spintronics, ferroelectrics

The Anomalous Hall Effect (AHE) in ferromagnetic metals and Spin Hall Effect (SHE) in nonmagnetic materials have attracted a renewed interest in the last decades. One can notice that AHE and SHE have the same origin, namely spin-orbit interaction in the presence of magnetic ordering for AHE and without magnetic ordering for SHE. Detailed analyses of the mechanisms responsible for these two effects may be found in reviews [1–3]. These mechanisms are divided into two groups: intrinsic ones and extrinsic ones. The former appear in pure metals and have topological nature, closely connected with Berry curvature. Extrinsic mechanisms are due to asymmetric electron scattering on defects in presence of spin-orbit interaction. Two main types of scattering are considered: skew scattering [4, 5] and side-jump scattering [6]. Most of theoretical papers on AHE and SHE considered the case of infinite homogeneous samples. References [7, 8] also investigated AHE for multilayers and for highly inhomogeneous media.

Let’s consider a magnetic tunnel junctions (i.e. a sandwich of two ferromagnetic layers separated by a dielectric spacer, MTJ – Magnetic Tunnel Junction) (Fig.1) submitted to a bias voltage applied between the two F-electrodes supposed to be made of the same ferromagnetic material. In this study, we are primarily interested by the Hall voltage which may appear between the opposite sides of the tunnel barrier due to the Hall current inside the spacer in presence of spin-orbit scattering on impurities. We will show that these Hall and spin Hall current do exist and that moreover, for the antiparallel orientation of the magnetizations in the two ferromagnetic layers, a spontaneous transverse Hall voltage exists, even in the absence of any applied bias voltage.

The Hall currents were calculated using Keldysh formalism [9]. The electrons were described as forming a free electron gas submitted to s-d exchange interaction. As an example, the Green functions for the considered system (Fig.1) and for z-projection of electron’s spin antiparallel to the magnetization in the left electrode are:

\[
G_{\text{AP},x\rightarrow x'}^{z}(r, r') = \frac{1}{N} \sum_{n \epsilon} \frac{1}{2q^2D} e^{i\kappa(t)(y-y')} e^{i\kappa(t)(z-z')} \\
\times \left( e^{q(x-x')} (q+ik_2) + e^{-q(x-x')} (q-ik_2) \right) \\
\times \left( e^{q(x'-x)} (q-ik_1) + e^{-q(x'-x)} (q+ik_1) \right),
\]

FIG. 1: Schematic illustration of MTJ. F - ferromagnetic layers, O - insulating spacer. Arrows denote the direction of magnetizations in electrodes, for parallel (P) and antiparallel (AP) orientations. Inset schematically illustrates the dependence of density of states (ν - in arbitrary units) of spin up tunnelling electrons on the distance from the interface for P and AP orientations.
\[ G_{AP,x<\pi}(r, r') = \frac{1}{N} \sum_{\pi} \frac{1}{2qD} e^{i\pi(x-y')} e^{i\pi(z-z')} \times \left( e^{i\psi(x-x_1)}(q - ik_1) + e^{-i\psi(x-x_2)}(q + ik_1) \right) \times \left( e^{i\psi(x'-x_2)}(q + ik_2) + e^{-i\psi(x'-x_2)}(q - ik_2) \right), \] \hspace{1cm} (2)

where:

\[ \mathcal{D} = \left( e^{i\psi}(q - ik_1)(q - ik_2) - e^{-i\psi}(q + ik_1)(q + ik_2) \right), \]

\[ q = \sqrt{\frac{2m}{\hbar^2}(U - E) + \kappa^2}, \]

\[ k_{1,2} = \sqrt{\frac{2m}{\hbar^2}(E \pm J_{sd}) - \kappa^2}. \]

In [1] and [2] “AP” means the antiparallel orientation of magnetizations in the two ferromagnetic electrodes and \( x_1, x_2 \) are the ferromagnetic/insulator interfaces coordinates. In [3], \( U \) is the barrier height, \( E \) is the electron energy, \( J_{sd} \) is s-d exchange energy. For the opposite direction of spin, all projection changes in (1) and (2) are straightforward. From [1] and [2], it follows that for the considered system, a finite density of states exists in the energy gap within the barrier due to proximity effect, which decreases exponentially with the distance from F/O interfaces (Fig[1]). In other words, a quasi-two-dimensional electron gas exists inside the barrier near the interfaces. Similarly to three dimensional topological insulator, this electron gas can give birth to charge and spin currents [10]. Evidently the mechanisms of creation of these surface states are different in the two cases. Let’s suppose now that the tunnelling electrons experience scattering on impurities with spin-orbit interaction. This asymmetric scattering deviates the electrons in the direction perpendicular to the tunnel current and to the projection of their spin. So if the current is spin-polarized, a Hall voltage appears transversally to the tunnel barrier. Quite interestingly, in antiparallel magnetic configuration of the MTJ, this AHE appears spontaneously even in the absence of bias voltage across the tunnel barrier. In addition, as illustrated in Fig[2], if the two ferromagnetic materials were assumed to have different spin-polarizations, a spontaneous spin unbalance (spin Hall effect) would also appear between the two transversal sides of the tunnel barrier at zero bias voltage. This would even be true if one of the ferromagnetic electrode was replaced by a non-magnetic metallic electrode.

To investigate this effect we added into the free electron Hamiltonian, the impurity potential including spin-orbit interaction and calculated the induced perturbation to the wave functions:

\[ \psi = \psi_0(r) + \int G(r, r') V_{so}(r') d^3r' = \]

\[ \psi_0(r) + \int \delta(r - r_1)(a_0^* \lambda_0) d^3r' \times \left[ G(r, r') i\sigma_z \left( \frac{\partial}{\partial x} \frac{\partial}{\partial y'} - \frac{\partial}{\partial y} \frac{\partial}{\partial x'} \right) \psi_0(r') \right]. \hspace{1cm} (4) \]

In [4] \( \lambda_0 \) represents the intensity of spin-orbit interaction, \( a_0 \) – lattice parameter, \( r_1 \) – position of the impurity, \( \sigma_z \) – z-component of Pauli matrix. Zero order wave function for the left-to-right and right-to-left tunnelling electrons are correspondently:

\[ \psi_{AP,l}^\dagger = \frac{2\sqrt{k_1}}{\mathcal{D}} \times \left( e^{i\psi(x-x_2)}(q + ik_2) + e^{-i\psi(x-x_2)}(q - ik_2) \right), \hspace{1cm} (5) \]

\[ \psi_{AP,r}^\dagger = \frac{2\sqrt{k_2}}{\mathcal{D}} \times \left( e^{i\psi(x-x_1)}(q - ik_1) + e^{-i\psi(x-x_1)}(q + ik_1) \right). \hspace{1cm} (6) \]

Now it is easy to calculate the Hall current in ballistic regime in the first order on spin-orbit interaction:

\[ j_H^z = \frac{e}{2\pi \hbar} \int \frac{f(E)}{(2\pi)^2} dE \]

\[ \times \int i\sigma_z \left( \psi_i^\sigma \frac{\partial}{\partial y} \psi_i^\sigma - \psi_i^\sigma \frac{\partial}{\partial y} \psi_i^\sigma \right) \left( \frac{E}{(2\pi)^2} \right) d\mathcal{D} \]

\[ \times \int i\sigma_z \left( \psi_r^\sigma \frac{\partial}{\partial y} \psi_r^\sigma - \psi_r^\sigma \frac{\partial}{\partial y} \psi_r^\sigma \right) \left( \frac{E}{(2\pi)^2} \right) d\mathcal{D}, \hspace{1cm} (7) \]

where \( f(E) \) – Fermi distribution for the left electrode, \( f(E + eV) \) – the same for the right one, \( V \) – applied voltage. Subscript “(…)” denotes the first order terms on spin-orbit interaction in the expression in brackets.
Substituting (4), (5) and (6) into (7) and averaging on the position of impurities \( r_i \) yields the following expressions for the spin-up all current originating respectively from left (\( l \)) and right (\( r \)) electrodes in AP configuration:

\[
\begin{align*}
&j_{AP, l}^\uparrow = \int d\kappa_x d\kappa_z dE \frac{4\lambda_0 \kappa_y^2 k_1}{|\mathcal{D}|^4} f(E) \\
&\times \left[ (e^{2q(x-x_z)} - e^{-2q(x-x_z)} - e^{-2q(x-x_z) - e^{2q(x-x_z)}}) (q^2 + k_y^2)^2 \\
&+ 2(e^{2q(x-x_z)} - e^{-2q(x-x_z)}) (q^2 - k_y^2) (k_z^2 - k_y^2) \\
&+ (e^{2q(x-x_z)} - e^{-2q(x-x_z)}) \\
&\times (q^2 + k_y^2)((q^2 + k_z^2) - (k_y^2 - k_z^2)) \\
&- 2(e^{2q(x-x_z)} - e^{-2q(x-x_z)}) \\
&\times ((q^2 - k_y k_z) (q^2 - k_z^2)) \right],
\end{align*}
\]

\[
\begin{align*}
&j_{AP, r}^\uparrow = \int d\kappa_x d\kappa_z dE \frac{4\lambda_0 \kappa_y^2 k_2}{|\mathcal{D}|^4} f(E + eV) \\
&\times \left[ (e^{2q(x-x_z)} - e^{-2q(x-x_z)} - e^{-2q(x-x_z) - e^{2q(x-x_z)}}) (q^2 + k_y^2)^2 \\
&+ 2(e^{2q(x-x_z)} - e^{-2q(x-x_z)}) (q^2 - k_y^2) (k_z^2 - k_y^2) \\
&+ (e^{2q(x-x_z)} - e^{-2q(x-x_z)}) \\
&\times (q^2 + k_y^2)((q^2 + k_z^2) - (k_y^2 - k_z^2)) \\
&- 2(e^{2q(x-x_z)} - e^{-2q(x-x_z)}) \\
&\times ((q^2 - k_y k_z) (q^2 - k_z^2)) \right].
\end{align*}
\]

In the limit \( e^{-2q x} < 1 \), in parallel configuration of the MTJ:

\[
\begin{align*}
&\langle j_{l+r}^{\uparrow+4} \rangle_{\text{AHE}}^{P, \text{skew}} = \frac{4}{15\pi} \frac{e^2}{2\pi \hbar} \frac{\tilde{\lambda} c}{U b} (E_F^\uparrow k_F^\uparrow - E_F^\downarrow k_F^\downarrow), \\
&\langle j_{l+r}^{\uparrow+4} \rangle_{\text{SHE}}^{P, \text{skew}} = \frac{4}{15\pi} \frac{e^2}{2\pi \hbar} \frac{\tilde{\lambda} c}{U b} (E_F^\uparrow k_F^\uparrow + E_F^\downarrow k_F^\downarrow),
\end{align*}
\]

and in antiparallel configuration:

\[
\begin{align*}
&\langle j_{l+r}^{\uparrow+4} \rangle_{\text{AHE}}^{\text{AP, skew}} = \frac{8}{105\pi} \frac{e}{2\pi \hbar} \frac{\tilde{\lambda} c}{U b} (E_F^\uparrow k_F^\uparrow + E_F^\downarrow k_F^\downarrow), \\
&\langle j_{l+r}^{\uparrow+4} \rangle_{\text{SHE}}^{\text{AP, skew}} = \langle j_{l+r}^{\uparrow+4} \rangle_{\text{SHE}},
\end{align*}
\]

where \( \lambda = 2m a_0^2 \lambda_0 / h^2 \) – dimensionless constant of spin-orbit interaction, \( c \) – atomic concentration of impurities.

One may notice that in contrast to the tunnelling current through the tunnel barrier, the expressions of the Hall and spin Hall currents do not contain the small parameter \( e^{-2q x} \). Instead, the averaged Hall voltage decreases inversely proportional to the barrier thickness. Its amplitude is proportional to the small parameter \( \lambda_0 \) related to the intensity of the spin-orbit interaction. The absence of \( e^{-2q x} \) in the expression for \( j_{l+r}^{\uparrow+4} \) further indicates that this predicted Hall and spin Hall effects have a surface nature in contrast to the tunnelling current.

Up to now, the case of “skew” scattering was considered. In addition to this scattering mechanism, another contribution to Hall and spin Hall currents originates from another term in the operator of quantum mechanical velocity, proportional to spin-orbit interaction:

\[
\dot{\mathbf{v}} = \frac{\hbar}{m} \mathbf{r} - i [\mathbf{r} \times \mathbf{H}] = \frac{\hbar k}{m} + \lambda [\mathbf{\sigma} \times \nabla V(r)],
\]

where \( V(r) \) – potential of impurity, \( \lambda \) – spin-orbit constant. This additional contribution to the Hall current is equivalent to a “side jump” mechanism [1]. In the present case it is written in final form as:

\[
\begin{align*}
&\langle j_{l+r}^{\uparrow+4} \rangle_{\text{AHE}}^{P, \text{skew}} = \frac{4}{15\pi} \frac{e^2}{2\pi \hbar} \frac{\tilde{\lambda} c}{U b} (E_F^\uparrow k_F^\uparrow - E_F^\downarrow k_F^\downarrow), \\
&\langle j_{l+r}^{\uparrow+4} \rangle_{\text{SHE}}^{P, \text{skew}} = \frac{4}{15\pi} \frac{e^2}{2\pi \hbar} \frac{\tilde{\lambda} c}{U b} (E_F^\uparrow k_F^\uparrow + E_F^\downarrow k_F^\downarrow),
\end{align*}
\]

Next the obtained expressions for Hall currents and spin Hall currents were averaged over the coordinate \( \mathbf{r} \) and integration over momentum \( \vec{\kappa} \) and energy \( E \) yields
\[ \langle j_{l+r} \rangle_{\text{AP,sj}}^{\text{SHE}} = \langle j_{l+r} \rangle_{\text{P,sj}}^{\text{SHE}} \]

First of all, we note that both contributions into the Hall and spin Hall currents are proportional to the concentration of impurities. This contrasts to the usual Hall conductivity in ferromagnetic metals which is inversely proportional to this concentration for the skew scattering and does not depend on concentration for the side jump mechanism. However in the present case, Hall current in metallic ferromagnetic electrodes is proportional to the current in this electrode, itself proportional to the small parameter \( e^{-2q_\theta} \). Therefore, for thick enough insulating spacer, Hall and spin Hall effects inside the spacer may become much larger than the corresponding effects within the ferromagnetic electrodes.

To find the Hall voltage \( V_H \), we divided the expressions for Hall current by conductance in \( y \)-direction:

\[
G = \frac{e^2}{2\pi \hbar} b \sqrt{\frac{2m}{\hbar^2} U} \\
\times \left[ \left( 1 - \sqrt{1 - \frac{E_F^+}{U}} \right) + \left( 1 - \sqrt{1 - \frac{E_F^-}{U}} \right) \right].
\]

Estimated value of \( V_H \) is \((10^{-5} \text{ to } 10^{-3}) V\), for \( \lambda \) in interval \((10^{-2} \text{ to } 10^{-1}) \) and \( c \) in interval \((0.01 \text{ to } 0.1) \). The most interesting conclusion is the existence of a Hall voltage for AP-configuration even in absence of any applied voltage. It means that an insulating spacer sandwiched between two ferromagnetic electrodes in AP-configuration exhibits a spontaneous electric polarization i.e. a spontaneous transverse ferroelectricity due to proximity effect. The latter results from the asymmetric scattering on spin-orbit impurities of tunnelling electrons penetrating into the insulating barrier from the ferromagnetic electrodes.

To experimentally measure this effect, one possibility would be to make electrically isolated metallic islands aside of the tunnel barrier. These islands would get charged by electrostatic influence with the charges arising on the side walls of the tunnel barrier. Measuring the voltage between these islands and the MTJ electrodes in parallel and antiparallel magnetic configuration with an electrometer could allow to detect and measure this new phenomenon of spontaneous transverse ferroelectricity in MTJ.

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