Loss of spin polarization in ferromagnet/ferroelectric tunnel junctions due to screening effects

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Electric field control of magnetization allows further miniaturization of integrated circuits for binary bit processing and data storage as it eliminates the need for bulky sophisticated systems to induce magnetic fields. Magnetoelectric coupling inherent to the bulk of multiferroic films or control of spin orientation in magnetic layers via piezoelectric strain in dual component composites have been two approaches standing out. Another magnetoelectric effect is spin-dependent screening that occurs at dielectric/ferromagnet interfaces which is of great importance for spin selective tunnel junctions. Here, we analyze the spin-dependent screening of ferroelectric polarization in a film interfacing ferromagnetic electrodes using the continuity equations in continuum media. The competition between the electrostatic and the magnetochemical potential in the FM electrodes gives rise to a reduction in the net magnetic moment near the interface due to spin mixing, extending to a distance comparable to the Thomas-Fermi screening length. Our continuum media treatment shows that the local spin population in spin subbands near the interfaces can dramatically deviate from bulk, which is in qualitative agreement with recent first principles results. We compute the tunneling currents for the majority and minority spins using the Wentzel-Kramers-Brillouin approximation as a function of ferroelectric polarization. We find that the spin polarization tends to disappear for increasing values of ferroelectric polarization in direct connection with the increase in subband spin population for minority spins at the interface.

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1. Introduction

Magnetoelectric coupling in bulk of perovskite oxides has been a forthcoming topic of interest since the observation of this phenomena near or above room temperature in ABO_3 type oxides. A is usually a rare earth and B is a transition metal element hosting spins when in the elemental state with O sites aiding in double-exchange between B sites generating long range magnetic ordering. The manifestation of the Dzyhalozhinski-Moriya coupling in some rare earth based perovskites [1-4] has paved the way to prescribe compositions that would allow coexistence of magnetic and electrical ordering in the bulk of a number of cubic perovskite materials. The symmetry correlation between the spontaneous electrical dipole order and the magnetic dipoles in the unit cell of such materials has allowed manipulating one order parameter via control of the other[5, 6]. From a technological point of view, control of the magnetic order through electric fields has been rather attractive as this would eliminate bulky components required to generate magnetic fields [7-12]. Several magnetoelectric oxides manifest magnetism by mechanisms such as double-exchange between B-sites via oxygen sites and is of ionic origin [13-22] that give rise to a small but finite ferroelectric (FE) polarization, namely "improper ferroelectricity" [23]. In fact, many material systems investigated in the past decade under the title of multiferroic behavior fall into this category. Dual phase systems where the material is microscopically engineered to exhibit two distinct phases with each phase having its own order parameter have also been on the agenda of research groups in attempts to create extrinsic multiferroicity [24-35]. These systems usually rely on the strain sensitivity of both magnetic and electrical order parameters in the distinct phases. Inducing strain in one component via an electric field that alters spin alignment in the magnetic phase via magnetostriction has been the key parameter to generate the magnetoelectric coupling between the two order parameters in a composite-like dual phase system.

Another type of magnetoelectric coupling that is inherent to dielectric/FM interfaces occurs due to spin dependent screening. Contrary to the numerous efforts focusing on multiferroic compositions and composites, there are a handful of works that focus on this mechanism at dielectric/ferromagnet (DE/FM) interfaces [36-38]. Upon application of a bias to a contact between a DE and a FM conductor, the screening of the polarization charges induced on the dielectric side of the interface becomes spin dependent. This naturally occurs because carriers accumulating or depleting the interface on the FM side are subject to the exchange field, hence the subband density of states (DOS) differ for spins of opposite sign. The DOS for the up and down spins in the valence band of the FM are shifted along the energy axis by an amount proportional to the strength of the internal exchange field whose origins are quantum mechanical. This situation can be altered by the polarization charges from the DE side when under bias and can be quite significant especially in thin film junctions where the screening lengths can become comparable to film thickness. Thus, a relatively high bias applied to a dielectric/FM junction can generate strong changes in the interface magnetization of the FM, either enhancing it or degrading it depending on whether accumulation or depletion of spins occurs due to local electrostatics. It has been reported that a very small change occurs in the interface magnetization of metallic FMs [39] as the screening lengths are very short owing to the very large density of states near the Fermi level, for instance in 3d metals. In FM oxides, on the other hand, that recently attracted great attention, changes amounting to 50% or more in the interface magnetization at distance of about 10 nm or more have been reported [40-42]. The main mechanism of such dramatic changes in interface magnetization of FM oxides can be shown to be purely due to the accumulation/depletion behavior of a doped semiconductor with the only difference that the carriers are spin polarized.

Spin accumulation and depletion at interfaces through the application of a bias via the mechanism mentioned above forms the basis on which magnetoresistive junctions operate, in particular, the junctions where the tunnel magnetoresistive (TMR) effect is tailored [43-52]. In conventional FM/DE/FM TMR stacks, one can obtain spin polarized tunnelling currents that are determined by the spin states of electrons in the FM electrodes. TMR junctions consist of two FMs separated by a thin layer of dielectric, a corollary of the Giant Magnetoresistive (GMR) stack. The spin polarization and magnitude of currents across a TMR stack depends on the relative orientation of the magnetism in the FM electrodes and a bias simply controls the electrical barrier to spin tunneling via the polarization of the dielectric. Shortly after the demonstration of the TMR effect in FM/DE/FM thin film stacks, replacing the DE with a ferroelectric (FE) layer entered the agenda of the scientific groups working on the development of devices for spintronics. The driving force behind such a pursuit was that the FE polarization can dramatically alter the on/off ratios of spin currents depending on the direction of remnant dipoles as they can easily be switched under a few volts of bias. FE TJs sandwiched between metal and semiconductor electrodes have already been proven to generate on/off ratios reaching 10^3-10^5 [53-59].

Inspired by results from to metal/FE/semiconductor junctions, a distinct control of spin currents in a gate-like setting via the FE layer appears to be an attractive option due to the ratio of the on/off states of spin currents that can vary by several orders of magnitude [35, 60-62]. TMR in FE TJs has been studied but similar to the TMR effect in FM/DE/FM stacks with the DE as the TJ, loss of spin polarization in tunnelling currents at moderate to high bias values is a persisting problem [63-65] even in the case of magnetization being parallel in the FM electrodes. The origin~~s~~ of such an outcome has been discussed extensively by a few authors for DE TJs. [48, 66-68] The flipping of the spins of "hot" electrons (those who have gained energy above the Fermi level of the FM electrodes) following tunnelling, magnon excitations and scattering events from defects inside the TJ that induce spin flips were discussed as major scenarios degrading the TMR effect. The changes in interface states in TJ/FM junctions has been mentioned in a few works [36, 68]. From the continuum media perspective one would expect a competition between electrostatic and magnetochemical potential an electron feels near a dielectric/FM interface under bias, keeping in mind magnetochemical potential pushes minority carriers to higher energies (some of which flip their spins and become majority carriers) via band shifts and charge distribution occurs always to minimize electrostatic energy. The finite penetration of the electric field to the surface of a metallic FM in a dielectric/FM junction under a potential drop will mostly be screened by majority spin electrons of the FM near the Fermi level. Strong electric fields could require carrier densities much greater than the population density allowed by the subband DOS of the majority spins. It can thus be expected that minority spin carriers can take effect and participate in the screening process as long as the energy difference between the spin subbands is not extreme such as in the case of half-metals. FEs can generate very strong fields near a metallic or semiconductor interface and pave a way to effectively manipulate carriers as well as their spins.

Here, we aim to understand the effect of the electrostatic screening process of FE polarization charges on magnetoresistive effects. For this purpose, we study a FM/FE/FM stack using the continuity equations in continuum media and study the spin dependent screening at the FM interfaces that has important implications for obtaining the TMR effect from such structures. The competition between the electrostatic and the magnetochemical potential of carriers at the FE/FM interfaces is demonstrated. Spin mixing, namely reduction of the majority/minority spin population ratio, could occur at the interfaces where the local density of states (LDOS) for minority spins is greater than in that of the bulk. This is driven by the need to screen polarization charges if the FE layer has a relatively strong polarization or is under a strong bias. By considering majority and minority spin channels, we try to quantify the limits of FE polarization and applied bias beyond which spin polarized currents are unlikely that would result in reduction of the TMR. We prove so by directly computing the tunnelling currents for majority and minority spin channels using the Wentzel-Brillouin-Kramers (WKB) approximation and discuss the results in the light of barrier heights for the two spin channels.