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Spin transport in metal and oxide devices at the nanoscale

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Abstract

Here we discuss a non-destructive technique that characterizes spin and charge transport at the nanometer scale, across buried layers and interfaces, in magnetic memory elements as used in spin transfer torque based Magnetic Random Access Memory (STT-MRAM). While probing in the current-perpendicular-to-plane direction, this method enables quantification of essential spin transport parameters as length and time scale, spin polarization in buried layers and interfaces, visualization of domain wall evolution across buried interfaces, besides investigating the homogeneity of transport, at the nanoscale, in spintronics devices.

Introduction

Spintronics is an emerging field of science and technology that utilizes the electron spin for significantly enhanced or fundamentally new device functionality. Following the discovery of the giant magneto resistance effect (1) more than a decade ago, this field has now witnessed a significant revolution, encompassing new materials (organic materials, graphene, graphite, complex oxide materials as multiferroics, manganites,) and new physical phenomena. Many of the beyond-CMOS devices as the STT-MRAM, spin transistors, hybrid devices of ferroelectric (or ferromagnetic)/semiconductor or spin based devices based on emerging materials poses several challenges related to the modification of the spin and charge transport when downsized to the nanometer scale. Thus, it is relevant to look for characterization methods to probe and understand spin transport in such devices at the nanoscale. Several characterization techniques related to either qualitative structural analysis, or static and dynamic imaging of domains in either ferromagnetic or ferroelectric materials exists today along with other scattering probes based on neutrons, x-rays, electrons, photons etc. However, a non-destructive method which combines qualitative structural analysis while yielding quantitative information of spin (charge) transport parameters with nanometer resolution along with the simultaneous visualization and imaging of electronic transport across buried layers is lacking. Such insights will be essential for exploiting the potential of spintronic devices based on emerging materials when downsized to the nanometer scale. Here we show that by using a three-terminal

transistor configuration (Fig.1 and 2) as in the Ballistic Electron Emission/Magnetic Microscopy (BEEM/BEMM) (2) we can probe (sub-surface) charge/spin transport and local magnetic switching in novel magnetic heterostructures and at an energy regime where non-equilibrium processes, relevant for the operation of such devices, are prevalent. In BEMM, unpolarized electrons are injected perpendicular to the device layers through a thin tunnel barrier, at typical energies between 0.6-2 eV above the Fermi energy (E_F). Hot electrons in the BEMM configuration are injected from the STM (Scanning Tunneling Microscope) tip across a tunnel barrier into the base where transport and attenuation occur due to a combination of scattering processes followed by propagation to the metal/semiconductor collector (Schottky) interface, which serves both as an energy and momentum filter. At such energies, for electron transport in the thin base layers, scattering processes include inelastic scattering with large energy losses, elastic scattering at defects and grain boundaries leading to a redistribution of electrons over different momentum states, spin wave scattering and other scattering processes that occur typically within 10 fs of electron transit through such thin layers. The collected current also depends on the overlap of the electronic states (both energy and momentum) at the metal/semiconductor interface, their effective masses and defined by a critical angle of collection at this interface. Depending on the energy and velocity component of the transmitted electrons, perpendicular to the Schottky interface, this critical angle is typically between 2° - 10° ; electrons that scatter at angles larger than this cone will not be collected. Transport at these energies is characterized by an important length scale, known as the attenuation length, λ , which depends on the product of the inelastic scattering time τ and group velocity v_g of the states into which the hot carriers propagate (3). Extraction of the attenuation length, λ , and studying its energy dependence in different metallic and ferromagnetic materials is essential to the design and optimization of hot electron based devices in spintronics. Further in BEMM, the injection and collection of hot electrons is local (~ 10 nm) and thus is also used to obtain spatially resolved information about hot electron transport. This is crucial to the understanding of local magnetic switching (4) in different ferromagnets in a spin valve stack when downsized to the nanometer scale. This

technique has also been used to study spin transport of both electrons and holes (3) which is relevant for complementary Spintronic device applications.

In recent years, devices that exploit hot electron transport have become increasingly popular in spintronics. This includes the spin valve transistor and the magnetic tunnel transistor (5), spin-transfer torque devices (6), Si spin injection devices (7), graphene based optoelectronic devices (8), manganite hot electron transistors (9) etc. Investigating the current-perpendicular-to plane transport of hot electrons at the nanoscale and the factors that govern, limit and characterize electron transport is essential to understand the performance of these devices. This is also relevant for emerging devices based on strongly correlated oxides, for which a comprehensive understanding of the electronic phase separation at such length scales is essential to the design and understanding of the device functionalities.

Spin dependent transport and magnetic resolution in metallic spin valves

A typical device structure used to study hot electron spin transport is shown in Fig.1b. Such spin valve structures are the essential ingredient of STT-MRAM and other spintronics based devices. Substrates consists of buffered hydrofluoric acid (HF)-etched *n(or p)*-Si(100) or *n(or p)*-Si(111) with a lithographically defined area of 150 μm diameter. This involves removal of the 300 nm dry SiO_2 followed by a sequence of photolithography steps to define the active area in the Si substrate. The Si surface is hydrogen terminated using 1% HF and thin magnetic layers are deposited using a thermal evaporator. The top layer is capped with a thin Au layer to prevent oxidation of the top magnetic layer. The bottom Cu layer forms a Schottky barrier with the Si substrate. Local scale spin transport in the presence of a magnetic field in a Si(100)/Cu(10 nm)/NiFe(4 nm)/Au(10 nm)/Co(4 nm)/Au(5 nm) device is shown in Fig 3. The temperature of operation in BEMM is determined by the junction resistance at the Schottky interface. For junction resistances $> 1\text{G}\Omega$ at zero bias, the transmitted hot electrons can be easily detected, otherwise the noise due to the diode dominates. Thus while a Si/Au Schottky interface enables BEEM measurements at RT ($\phi_B \sim 0.8\text{ eV}$) that using a Cu/Si interface necessitates the measurement temperature to be lower than RT ($\phi_B \sim 0.6\text{ eV}$). Clear difference in the hot electron transmission is observed for the parallel and antiparallel alignment of the magnetic spin valve when the *in situ* magnetic field is swept. To unambiguously demonstrate the spin dependence of transmission in the spin valve stack, magnetic hysteresis was recorded at the nanoscale ($\sim 10\text{ nm}$) while keeping the tip-sample bias and tunnel current fixed. This is done by sweeping the magnetic field and is recorded as the switching of the BEMM transmission from the parallel to the antiparallel state of the spin valve (Fig.3). We have

also used the potential of BEMM to obtain a spatial map of the collected current from buried layers and interfaces and at different magnetic fields. Evolution and characterization of magnetic domain walls at such interfaces is also done in this work. Further, application of the spin dependence of electron transport to high resolution magnetic imaging was demonstrated in yet another similar spin valve stack. The magnetic resolution was determined from the signal variation along a line crossing a 360° domain wall as shown in Fig 4. The signal reaches the transmission corresponding to the parallel state and the profile can be well described by a simple arctan function for a single bit transition (blue solid line). Defining the transition width as the distance between the points where 20% and 80% of the maximum signal are reached, the width is determined to be 16 nm. This is a first demonstration of such high (magnetic) resolution achieved by BEMM on metallic spin valves. It is worth mentioning that the resolution of the technique is not limited to this value but depends on the magnetic structures to be resolved at such energies and the associated layer stacks.

Oxide electronics

Complex oxide heterointerfaces are ideal platforms to study novel electronic properties that stem from strong electron correlation effects between the electron's spin, charge and orbital degrees of freedom. Memory and logic device concepts with these emerging materials will need to take into account the unconventional behaviour of the dielectric constant at such heterointerfaces, hysteretic change in resistance states due to current or voltage pulses, coupling of electric and magnetic domains etc. A generic feature of such complex oxides is the coexistence of several competing states leading to a complicated phase diagram. The observation of novel and unparalleled physical phenomena (10) ranging from metallic, superconducting or magnetic at well-defined interfaces between two complex oxide insulators has created tremendous fundamental interest. These materials have a principal advantage in device scaling because such heterostructures have metallic electron densities (10^{22} to 10^{23} cm^{-3}) even in their insulating state. This can play a significant role in circumventing the important bottleneck of performance limitation in miniaturizing conventional microelectronic devices due to quantum mechanical effects. Such interfaces are also attractive for designing devices that do not necessarily scale according to Moore's law. However, the challenge here is to understand the role of defects, strain and doping inhomogeneity, internal electric fields to charge and spin transport across such interfaces. We study this using the technique of BEEM using a Schottky interface between an unconventional semiconductor as doped SrTiO_3 and complex oxides as a manganite, a multiferroic as BiFeO_3 , an oxide metal as

SrRuO₃ ec. In this context, it can be mentioned that quite recently (9), a heteroepitaxial perovskite metal-base transistor using La_{0.7}Sr_{0.3}MnO₃ on an n-type oxide semiconductor as Nb doped SrTiO₃ has been demonstrated. While the characteristics of such devices ranges from hot electron to permeable base transistors, undoubtedly such devices open up new possibilities to study novel electronic phases in such materials. We have recently demonstrated hot electron transmission, at the nanoscale in thin layers of La_{0.7}Sr_{0.3}MnO₃ (LSMO) on Nb doped SrTiO₃ (STO) substrates. Using the technique of BEEM as shown in Fig.5, we have investigated electron transport in a current-perpendicular-to the plane of the device at different energies and studied the influence of strong correlation to electron transport. On singly terminated (TiO₂ termination) and well-characterized Nb:SrTiO₃ substrates (11), LSMO thin films of varying thickness (Fig.6) were deposited using Pulsed Laser Deposition and fabricated into diodes using standard UV lithography. Macroscopic electrical measurements of the interfaces show good rectifying properties with low reverse leakage currents. Using BEEM we find that the hot electron transmission in such epitaxial heterostructures depends strongly on the energy and momentum conservation and the local transmission probability at such interfaces. From the obtained BEEM spectra, a spatial distribution of Schottky Barrier heights as well as the BEEM transmission has been obtained (Fig.7). Temperature dependent measurements indicate the change in the energy band line up at the interface. Our experiments give a first insight into the behaviour of hot electron transport at the nanoscale in oxide devices and the energy dependence enables us to quantitatively isolate different scattering mechanisms prevalent here.

Summary

In summary, we have demonstrated the capabilities of BEMM in characterizing (spin) electronic transport in a vertical geometry and its application in different magnetic multilayers. Such transport studies with nanometer resolution gives insights into the band alignment at the Schottky interface as well as the homogeneity of current transport across such interfaces. This is essential to understand and tailor the performance of devices based on emerging materials and to understand their suitability for memory and/or logic devices in this rapidly evolving field of spintronics.

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Fig.1a: Ballistic Electron Magnetic Microscopy set up: A PtIr tip is used to inject hot electrons in a spin valve (Ferromagnet/Normal Metal/Ferromagnet) grown on a Si/Metal Schottky interface. The hot electrons are injected after tunnelling through a vacuum tunnel barrier. The emitter bias is kept negative such that hot electrons are injected by tunnelling into the base. After transmission in the base, they are then collected in the conduction band of the n-type semiconductor. An external magnetic field is applied *in situ* to study the spin dependent transmission in the spin valve layers. When both the FM layers are aligned parallel, the collected current is larger. For antiparallel alignment the collected current is smaller.

Fig.1b: A typical layer stack. Here the spin valve constitutes of Co (4 nm)/Au (10 nm)/NiFe (4 nm) layer stacks, thermally evaporated on a Hydrogen terminated substrate Si (100) substrate. The top Au (5 nm) layer is used as a

capping layer for *ex situ* transfer of the device into the BEMM and prevents oxidation of the top FM layer.

Fig.2 Energy schematics of the BEMM:Unpolarized hot electrons are injected from the PtIr STM tip across a vacuum tunnel barrier. They are then polarized in the first FM layer and after transmission through the entire layer stack is collected in the conduction band of the semiconductor if they satisfy the necessary energy and momentum criteria at that interface. The Schottky barrier height at the metal/semiconductor interface is shown as ϕ_B .

Fig.3(Left) Electron current as a function of tip bias for a Si(100)/Cu (10 nm)/NiFe (4 nm)/Au(10 nm)/Co (4 nm)/Au(5 nm) spin valve for a tunnel current of 4 nA. The black curve corresponds to the collected current for the configuration when both Co and NiFe layers are aligned parallel and the red curve corresponds to the antiparallel configuration when the softer NiFe layer has reversed its magnetization. Each curve represents an average of over 120 spectra taken at different locations in the device. (Right) Magnetic hysteresis for the same spin valve layer recorded at -1.2 V and for 4 nA of tunnel current at the local scale. The change in the current between the parallel and antiparallel configuration corresponds to a magnetocurrent of $275 \pm 50\%$.

Fig.4 BEEM image of the device layer in Fig.1b taken at a magnetic field of 50 Oe (antiparallel alignment). The image is recorded at -1.4 V and 3 nA. Presence of 360° domain walls is clearly visible. Magnetic resolution is determined from the BEEM current variation along the white line as shown in the right where the profile is fitted by a simple arctan function for a single bit transition. The magnetic resolution for hot electrons is thus determined to be 16 nm.

Fig.5 For BEEM studies on oxide heterostructures, the sample constituted of a thin epitaxial film of $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$ (LSMO) deposited on an isostructural 0.01 wt % Nb doped SrTiO_3 (Nb:STO) substrate, using Pulsed Laser Deposition technique. A PtIr tip is used to locally inject electrons into the sample by tunneling between the tip and the LSMO surface. The electrons transmitted perpendicularly through the LSMO layer is collected in the Nb:STO with a third electrical contact.

Fig.6 Atomic Force Micrograph (AFM) of a chemically treated and annealed Nb doped SrTiO_3 substrate. Single terminated TiO_2 planes on the substrate surface are visible. The step height of 3.9 Å corresponds to the distance between two TiO_2 planes.

Fig.7: (Top) Energy dependence of the electron transmission in a LSMO (3 nm) layer on Nb:doped SrTiO_3 substrate. The sign of the current corresponds to electrons flowing from the LSMO layer to the Nb:STO and into the ohmic back contact. The collected current is an average of 50 different spectra collected at different regions, in approximately 4 devices. $T = 300$ K. (Bottom) The square root of the BEEM transmission is fitted to the Bell-Kaiser model to extract the local Schottky barrier height at the LSMO/Nb:STO interface. The Schottky barrier height thus extracted is 1.08 ± 0.02 eV.