Tunnel magnetoresistance due to spin accumulation in nonmagnetic nanoparticles and its potential applications

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Recent studies on spin accumulation and tunnel magnetoresistance (TMR) in nonmagnetic metal nanoparticles are reviewed, and a potential device application for magnetoresistive random access memories (MRAMs) is proposed, based on the unique magnetotransport properties in nonmagnetic nanoparticles.
Key words: nanoparticle, spin accumulation, tunnel magnetoresistance, MRAM

1. INTRODUCTION

Spin accumulation gives rise to a variety of phenomena and functionalities, such as electrical manipulation of magnetization [1] and reconfigurable logic [2], which are useful for the development of spin electronic devices [3]. Nanoparticles are the systems in which highly efficient spin accumulation is expected to occur because of their small volumes. In fact, experimental observations of remarkable magnetotransport phenomena due to spin accumulation in magnetic and nonmagnetic nanoparticles were reported so far [4-9], as well as important theoretical predictions [10-13]. In this paper, the concept of spin accumulation is explained for a simple case, and recent experiments on spin accumulation in "nonmagnetic" nanoparticles are briefly reviewed. Finally, a potential application of spin accumulation and consequent tunnel magnetoresistance (TMR) in nonmagnetic nanoparticles to magnetoresistive random access memories (MRAMs) is proposed.

2. SPIN ACCUMULATION IN NONMAGNETIC NANOPARTICLES

Although spin accumulation appears wherever electron current passes through an interface of materials with different spin polarizations of conduction electrons, this paper focuses on symmetric double tunnel junctions with a nonmagnetic island (center) electrode because they are simple and typical systems for efficient spin accumulation. First, we consider spin accumulation in conventional double tunnel junctions in which the Coulomb blockade effect does not occur. Fig. 1(a) illustrates a schematic structure of a double tunnel junction consisting of the stacking of ferromagnetic electrode / tunnel barrier / nonmagnetic island electrode / tunnel barrier / ferromagnetic electrode. Fig. 1(b) shows the densities of states corresponding to each electrode in the antiparallel configuration of the electrode magnetization, assuming that spin relaxation rate is negligibly small. Due to the spin dependent tunnel conductance and spin conservations, finite spin accumulation appears in the island electrode, i.e., the number of up spin electrons is increased in the island electrode because the up spin electron current is larger than the down spin one for the incoming electrons while the down spin electron current is larger than the up spin one for the outgoing electrons. In the parallel configuration of magnetization, on the other hand, spin accumulation does not appear because the ratio of up and down spin electrons (= spin polarization) is not different between the incoming and outgoing electrons. It is noted that the spin dependent tunnel conductance is approximately given as the product of the densities of states of the corresponding spin electrons between electrodes.

Next, we consider the use of a nanoparticle. When the island electrode in Fig. 1(a) is replaced by a nonmagnetic nanoparticle, as shown in Fig. 2(a), spin accumulation still appears and the Coulomb blockade effect is added to the electron transport. The Coulomb blockade effect is characterized by the electrical charging energy of the system, which is given by the

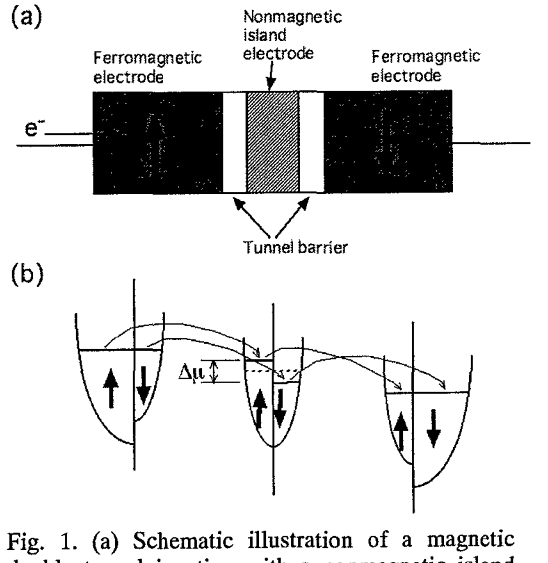

Fig. 1. (a) Schematic illustration of a magnetic double tunnel junction with a nonmagnetic island electrode, (b) spin resolved densities of states corresponding to each electrode.

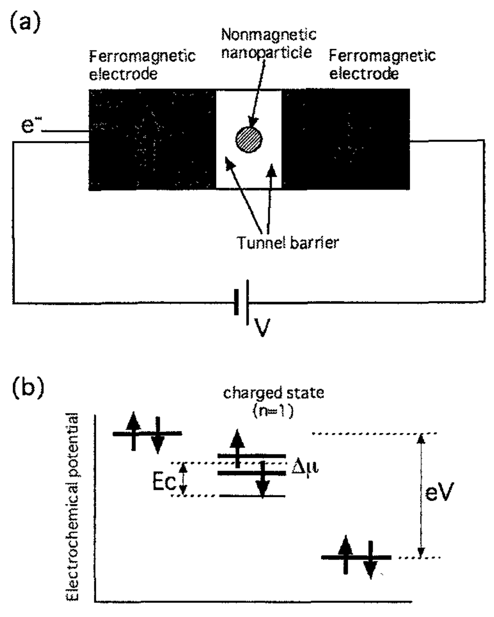

Fig. 2. (a) Schematic illustration of a magnetic double tunnel junction including a nonmagnetic nanoparticle as an island electrode, (b) an example of the energy diagram of the double tunnel junction including a nonmagnetic nanoparticle.

capacitance around the tunnel barrier between a ferromagnetic electrode and a nanoparticle. Due to the small volume of the nanoparticle, the capacitance C is quite small, resulting in the large charging energy Ec ($Ec = e^2/2C$ where e is the electronic charge), where the contribution of self capacitance of the nanoparticle is significant. For a nanoparticle of 1 nm in diameter, the charging energy is much larger than the thermal energy at room temperature (RT). Namely, the Coulomb blockade can be achieved even at RT for such a small particles. Nonmagnetic nanoparticles with the diameter of about 1 nm has been prepared in Au deposited on a MgO tunnel barrier [14].

The electron transport is prohibited, i.e. the Coulomb blockade occurs, unless voltage bias overcomes the charging energy. The energy diagram of the system helps us understand the transport properties including the Coulomb blockade state. As an example, Fig. 2(b) shows an energy diagram when the voltage bias is larger than the charging energy, and the charged state of the nanoparticle with one excess electron (n=1) contributes to the electron transport of the system. When the magnetization configuration is changed from parallel to antiparallel, spin accumulation appears in the nanoparticle and consequently the tunnel conductance of the system is modified. This modification due to the change in magnetization configuration leads to TMR even though the nanoparticle is nonmagnetic.

TMR due to spin accumulation in double tunnel junctions with a nonmagnetic island electrode is first reported in a semiconductor-based system. TMR of 38 % was observed at 4 K for a GaMnAs/AlAs/GaAs/AlAs/GaMnAs junction [15]. In metallic nanoparticles, Zhang *et al.* reported TMR of about 10 % at 4.2 K in Co/AlO/Al-nanoparticles/AlO/Co junctions of a millimeter scale although the Coulomb blockade was not clearly observed in the current-voltage characteristics [5]. Furthermore, interesting transport properties such as the Hanle effect and the asymmetric bias-voltage dependence of TMR were shown in their paper. A problem of their work might be the absence of clear Coulomb blockade. The observation of clear Coulomb blockade evidences that the contribution of TMR due to direct tunneling between two Co electrodes is much suppressed. Bernand-Mantel *et al.* successfully observed remarkable TMR associated with clear Coulomb blockade and Coulomb staircases in a Co/AlO/Au-nanoparticle/AlO/Co junction prepared by their unique method [6]. They employed a conductive tip nanoindentation technique to prepare a nanometric double junction including a single metallic nanoparticle as an island electrode. TMR of about 6 % was observed at 4 K, and its inverse sign was regarded as an evidence of spin accumulation in the Au nanoparticle. However, systematic measurements, e.g. bias-voltage dependence, have not been performed yet. Recently, we also observed TMR due to spin accumulation in Au nanoparticles by using a Fe/MgO/Au-nanoparticles/MgO/Fe monocrystalline junction which was microfabricated into a submicron-sized pillar [7]. TMR of 1-2 % was observed at 4.2 K and the bias-voltage dependence of TMR was discussed. However, the observed TMR at low bias-voltages included the contribution of direct tunneling between the top and bottom Fe electrodes, and it could not be determined exactly how large current was needed to induce considerable spin accumulation in Au nanoparticles. In the current status, further experimental studies are required to well understand the spin accumulation and consequent TMR in nonmagnetic metallic nanoparticles.

3. POTENTIAL APPLICATIONS TO MAGNETIC MEMORY DEVICES

The Coulomb blockade effect makes it possible to control the resistance of the system through the bias and gate voltage [10,13,16], and this functionality can be used for selecting memory cells in random access memory devices. In this section, we propose a potential application of TMR due to spin accumulation in nonmagnetic nanoparticles to magnetic random access memory devices.

Fig. 3(a) shows a schematic illustration of the memory cell structure proposed. An information is stored in the center free ferromagnetic layer as the direction of magnetization. The staking at the lower part consisting of free ferromagnetic layer / tunnel barrier / nonmagnetic nanoparticles / tunnel barrier / pinned ferromagnetic (reference) layer is the structure for reading the direction of the free layer magnetization. The stacking at the upper part consisting of pinned ferromagnetic (spin-injector) layer / metallic spacer layer or tunnel barrier / free ferromagnetic layer is the one for changing the direction of the free layer magnetization by injecting spin polarized current, i.e., for writing information. Spin transfer torque from spin polarized conduction electron current to local spins [17] is considered to be the main mechanism of the change in

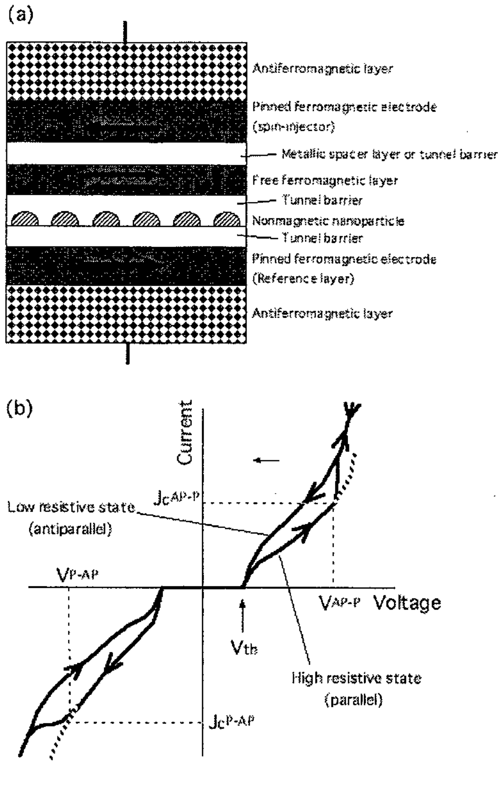

Fig. 3. (a) Schematic illustration of a memory cell using TMR due to spin accumulation and Coulomb blockade in nonmagnetic nanoparticles, (b) current-voltage characteristics of the memory cell structure.

the magnetization direction, while the contribution of spin accumulation to magnetization reversal in the free ferromagnetic layer may also be taken into account [18]. The top and bottom antiferromagnetic alloy layers fix the magnetization directions of two pinned layers, i.e., the spin-injector for magnetization reversal and the reference layer for detecting the TMR signal due to spin accumulation in the nonmagnetic nanoparticles.

The nonmagnetic nanoparticles are employed for the occurrence of the Coulomb blockade, and only the cell to which a bias-voltage exceeding the Coulomb threshold voltage V_{th} is applied becomes active for reading the information. As mentioned above, the diameter d of nanoparticles comparable to or smaller than about 1 nm is required for RT operations of the device since the electrical charging energy Ec of a nanoparticle, which is approximately given as $Ec = e^2/2C \sim e^2/4\pi\varepsilon'\varepsilon_0 d$ (ε_0: dielectric constant of vacuum, ε': relative dielectric constant of the barrier material) and $\varepsilon' \sim 10$ for typical barrier materials such as Al-oxide and Mg-oxides, should be about one order of magnitude larger than the energy scale of thermal fluctuation at RT.

The most important point in the proposed memory cell architecture is to use TMR due to spin accumulation in nonmagnetic nanoparticles instead of TMR in magnetic nanoparticles. This overcomes the problem of thermal fluctuation of magnetization which commonly emerges in applications of nanoparticles consisting of ferromagnetic materials. Namely, spin accumulation in nonmagnetic nanoparticles enable us to utilize both TMR and Coulomb blockade in nanoparticles.

Fig. 3(b) shows the current-voltage characteristics (*I-V* curve) in the memory cell shown in Fig. 3(a). A large spin polarized electron current from top to bottom of the cell structure makes the free layer magnetization direction switch to be antiparallel to that of the reference layer, while a large spin polarized electron current in the opposite direction leads to the parallel configuration of magnetization between the free and reference layers. Thus, the *I-V* curve shows a hysteretic behavior with the high and low resistance states.

Finally, let us discuss how long spin relaxation time in nanoparticles is required to realize this memory device. In order to maintain considerable spin accumulation in a nanoparticle, the interval time of successive tunneling events should be shorter than the spin relaxation time in the nanoparticle. Therefore, the condition for the appearance of spin accumulation is roughly given as $\tau_{sf} > e/i$ where i is the magnitude of reading current through a single nanoparticle. The validity of this relation is confirmed in previous studies based on numerical simulations [4,13]. Assuming that the cell size is reduced to be 100 nm x 100 nm for a high-density MRAM and that nonmagnetic nanoparticles with the number density of 4 x 10^{13} are used, one cell includes 400 nanoparticles and i is estimated to be j_c x $(100 \times 10^{-7})^2 / 400 \times (1/R_{RW}) = 1.25 \times 10^{-7}$ A for the current density of spin-polarized current induced magnetization reversal $j_c = 5 \times 10^6$ A/cm^2 and the current ratio between writing and reading $R_{RW} = 10$. τ_{sf} required is estimated to be $> e/i \sim 1$ fsec. Spin relaxation time in nonmagnetic materials satisfies $\tau_{sf} > \sim 1$ fsec [3], and furthermore a large enhancement of τ_{sf} has been found in nanoparticles [4].

4. SUMMARY

Recent studies on spin accumulation and TMR in nonmagnetic metal nanoparticles are reviewed. A potential MRAM cell application of TMR due to spin accumulation in nanoparticles is proposed, and it is shown that spin relaxation time in nonmagnetic materials is long enough to realize the proposed memory cell.

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