Current-induced tunnel magnetoresistance due to spin accumulation in Au nanoparticles

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Spin-dependent single electron tunneling was investigated in a magnetic double tunnel junction including Au nanoparticles as a center electrode. Tunnel magnetoresistance (TMR) clearly emerged with increasing spin-polarized current injected into Au nanoparticles and reached a maximum value of about 12% at 4.2 K. The observation indicates that spin accumulation occurs in Au nanoparticles and causes TMR. The spin relaxation time in Au nanoparticles, as estimated from the critical current for the appearance of TMR, is of the order of 10 ns, which is much longer than that in the bulk state. © 2008 American Institute of Physics. [DOI: 10.1063/1.2912036]

Spin accumulation is a nonequilibrium phenomenon occurring on the nanometer scale and besides being a basic concept in physics, it is one of the most important effects for spin electronic devices.[1] Spin accumulation in nanoparticles is of particular interest, since the particles's volumes are extremely small and, therefore, efficient spin accumulation is easily realized in principle. The interplay of single electron tunneling (SET) and spin accumulation[2–7] is expected to appear in magnetic SET structures incorporating nanoparticles that provide functionalities for controlling tunnel magnetoresistance (TMR) via the source-drain and/or gate voltages.[1,3,5,6] Moreover, spin accumulation in *nonmagnetic* nanoparticles has a unique merit in applications: nonzero TMR due to the spin accumulation in nonmagnetic nanoparticles occurs in spite of there being no spin polarization of conduction electrons in such nanoparticles and it is free from the superparamagnetic fluctuation of magnetization that affects ferromagnetic nanoparticle (nanograin) applications such as ultrahigh density magnetic recording media.

A few groups have reported TMR associated with spin accumulation in nonmagnetic nanoparticles.[8–11] Zhang *et al.*[8] observed TMR of about 10% and a Hanle effect at 4.2 K in double tunnel junctions of Co/Al–O/Al-nanoparticles/Al–O/Co, although the Coulomb blockade of Al nanoparticles was not so clear. Bernand–Mantel *et al.*[9] observed Coulomb staircases and TMR of about 7% at 4 K for a single Au nanoparticle by using their conductive tip nanoindentation technique. A change in the sign of TMR evidenced that the origin of TMR was not direct tunneling between ferromagnetic electrodes but spin accumulation in the Au nanoparticles. Theoretical calculations in the SET regime have shown that such an inverse TMR effect could appear for certain bias voltages and resistance ratios of the barriers.[6,11,12] However, more direct and physically important evidence for spin accumulation in a nonmagnetic nanoparticle in the SET regime is the observation of TMR emerging with increasing injection current.[4,6] This phenomenon is based on the fact that nonzero spin accumulation occurs when spin-polarized current becomes so

large that the time interval between tunneling events is shorter than the spin relaxation time. This behavior is indeed predicted by the theoretical calculation of Brataas *et al.*,[4] and the spin relaxation time τ_{sf} in a nonmagnetic nanoparticle can be estimated by a simple relation $\tau_{sf} \sim e/I_c^{TMR}$, where e is the absolute value of electron charge and I_c^{TMR} is the critical current for the appearance of TMR (e/I_c^{TMR}: time interval of tunneling events at I_c^{TMR}). In this study, we observed the evidence for spin accumulation, i.e., TMR emerging with increasing current, in a magnetic double tunnel junction with Au nanoparticles as a center electrode.

The sample structure of the double tunnel junction including Au nanoparticles as a center electrode is schematically illustrated in Fig. 1. Each layer was deposited at room temperature (RT) by molecular beam epitaxy and a MgO buffer and a Fe bottom electrode were postannealed at 600 and 400 °C, respectively. Epitaxial growth of each layer was confirmed by reflection high-energy electron diffraction, except for Au nanoparticles, since the diffraction intensity from the small Au nanoparticles was too weak to clearly observe. Formation of Au nanoparticles on a MgO(100) barrier layer was previously studied.[10,13] In the case of deposition of ~0.01 nm nominal thickness of Au, atomic force microscopy and scanning tunneling microscopy revealed that Au
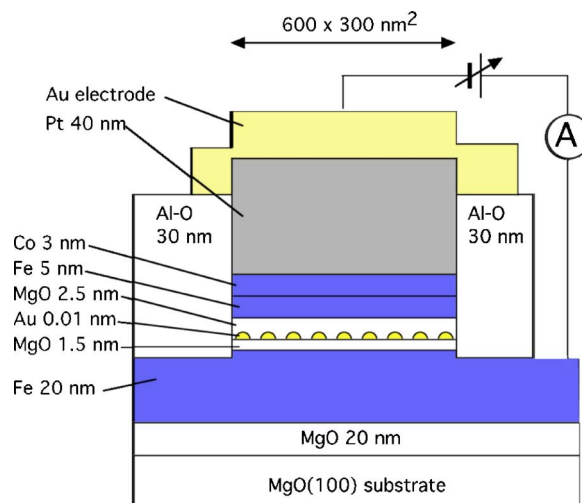
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FIG. 1. (Color online) Schematic illustration of a magnetic double tunnel junction including Au nanoparticles as a center electrode.
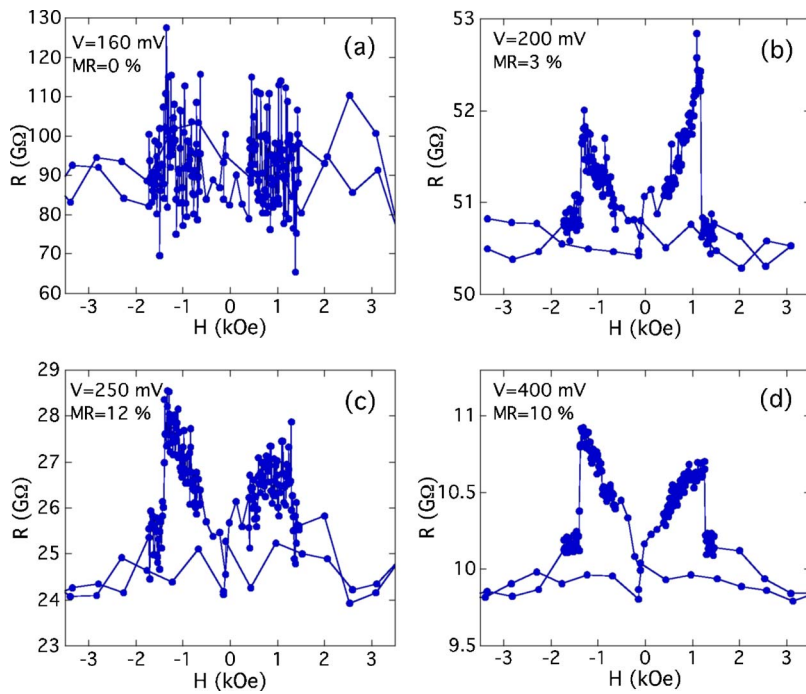
FIG. 2. (Color online) MR curves at 4.2 K and different bias voltages V for a magnetic double tunnel junction including Au nanoparticles; (a) $V=160$ mV, (b) $V=200$ mV, (c) $V=250$ mV, and (d) $V=400$ mV.

nanoparticles of 1–2 nm in diameter and $\sim 10^{12}$ cm^{-2} in number density were grown at RT. Microfabrication for the SET device structure shown in Fig. 1 was performed with electron beam lithography and Ar ion etching. A pillar measuring 600×300 nm^2 was made by Ar ion etching with a negative resist mask, and its sidewalls were covered by an Al–O insulating layer. Although the sample structure and the preparation conditions were similar to those for our previous samples,[10] we obtained samples with improved SET properties by adjusting the preparation conditions. Magnetotransport measurements were performed at 4.2 K by using an electrometer with an applied magnetic field of up to 7 kOe.

Figures 2(a)–2(d) show MR curves of the double tunnel junction at 4.2 K and different bias voltages (V). At $V=160$ mV, the sample resistance was significantly high (~ 100 G Ω) because of the effect of the Coulomb blockade of Au nanoparticles, and no apparent MR effect was observed. When V was increased to 200 mV, a small MR effect of about 3% appeared. The hysteresis of the MR curve indicates that the MR response arises in association with the change in the magnetization configuration of the top and bottom ferromagnetic metal electrodes, although complete antiparallel alignment of magnetization does not occur. With further increasing V to 250 mV, the MR ratio reached about 12%, showing a very similar MR hysteresis to the 200 mV case and lower sample resistance. In other words, the MR ratio clearly increased with increasing current. This behavior agrees with the characteristic of TMR induced by spin accumulation. At higher bias voltages, the MR ratio decreased, as shown in Fig. 1(d). A decrease in the MR ratio at high bias voltages is commonly observed for almost all TMR effects in a variety of systems. However, it is difficult to specify the mechanism.

Figure 3 shows the bias voltage dependence of the MR ratio together with the current-voltage (I-V) characteristics of the sample. The current is suppressed below $V=100$ mV, i.e., the Coulomb blockade region, and there is a small step at $V \sim 240$ mV, which presumably shows a Coulomb staircase. At higher bias voltages, many current channels may open

and therefore the Coulomb staircases become smeared out. The correlation between the I-V characteristics and the MR ratio indicates that the TMR effect emerges with increasing injection current into the Au nanoparticles. Note that the contribution of the leak current due to direct tunneling between top and bottom ferromagnetic electrodes is ruled out in the
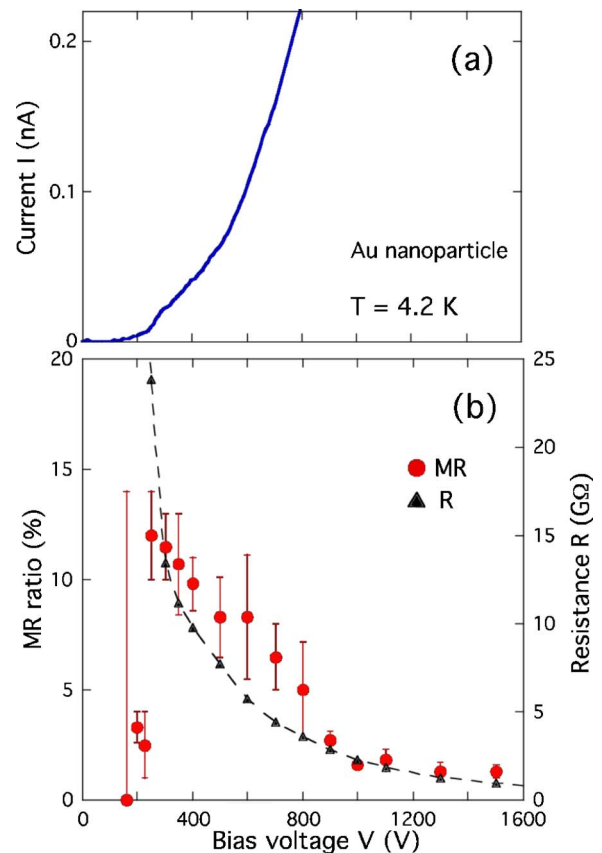

FIG. 3. (Color online) (a) Current-voltage characteristics and (b) MR ratio (closed circles) as a function of bias voltage at 4.2 K for a magnetic double tunnel junction including Au nanoparticles. The bias voltage dependence of tunnel resistance (open triangles) is also shown in (b).

observed TMR effect. Although the leak current may cause a nonzero TMR effect, the MR ratio due to leak current should monotonically increase with decreasing current through the nanoparticles, i.e., with decreasing bias voltage, because the current through the nanoparticles works as a shunt current against the leak current. This prediction for the case of leak current is in disagreement with the observation for $V < 250$ mV, and this disagreement means that the contribution of the leak current is negligible. In addition, a simple model analysis[14] makes it possible to evaluate the portion of the MR ratio due to the leak current at low bias voltages. By considering a parallel circuit consisting of dominant tunneling through nanoparticles and small direct tunneling between electrodes, a proportional relationship between the MR ratio due to leak current and the sample resistance R can be straightforwardly derived, i.e., MR ratio $\sim R$. If it is assumed that the observed TMR effects are caused by the leak current, the MR ratio due to leak current at $V = 160$ mV should be 60% from the observed MR ratios at $V > 250$ mV and R shown in Fig. 3, but this is too large to explain the observed MR ratio of $0 \pm 14\%$ at $V = 160$ mV.

Spin relaxation time τ_{sf} in the Au nanoparticles is estimated to be ~ 10 ns by using the relation $\tau_{sf} \sim e / I_c^{TMR}$ mentioned in the introduction and the experimental data in Figs. 3(a) and 3(b) ($I_c^{TMR} \sim 0.1$ nA). This is surprisingly longer than the spin relaxation time in the bulk state, which is ~ 10 ps, from the experiments of current-perpendicular-to-plane giant MR in FePt/Au/FePt nanopillars by Seki *et al.*[15] Although the enhancement of τ_{sf} seems unprecedentedly large, similar results have been reported.[7–9] For ferromagnetic Co nanoparticles, Yakushiji *et al.*[7] found that $\tau_{sf}(\sim 150$ ns$)$ is much longer than in the bulk state. For Au nanoparticles, a shorter limit of τ_{sf} can be derived from the data of the resistance and voltage in the MR curve reported by Bernand–Mantel *et al.*[9] In this case, τ_{sf} for Au nanoparticles is estimated to be > 5 ns. The experimental results of Zhang *et al.*[8] also suggest that τ_{sf} in Al nanoparticles is rather longer than in the bulk state. The enhancement of τ_{sf} in nanoparticles might be useful for future spin electronic applica-tions, although further studies are required to understand its mechanism.

In summary, we prepared a magnetic double tunnel junction including Au nanoparticles as a center electrode and it clearly showed magnetotransport properties in the SET regime. TMR emerged with increasing spin-polarized current injected into Au nanoparticles, showing that the observed TMR is caused by spin accumulation induced in Au nanoparticles. From the critical current for the appearance of TMR, τ_{sf} in a Au nanoparticle is estimated to be ~ 10 ns.

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