Temperature Evolution of Spin Relaxation in a NiFe/Cu Lateral Spin Valve

T. Kimura,1,2 T. Sato,1 and Y. Otani1,2

1Institute for Solid State Physics, University of Tokyo, 5-1-5 Kashiwanoha, Kashiwa, Chiba 277-8581, Japan
2RIKEN FRS, 2-1 Hirosawa, Wako, Saitama 351-0198, Japan

(Received 26 September 2007; published 13 February 2008)

Temperature dependence of spin relaxation process in a Cu wire has been studied by means of nonlocal spin-valve measurements. The spin-diffusion length of the Cu wire is found to take maximum at the characteristic temperature, below which the spin-diffusion length is reduced. The mechanism of the reduction can be explained by considering the spin-flip scattering due to the oxidized surface of the Cu wire. The thickness dependence of the characteristic temperature supports the interpretation with the surface oxidation.

DOI: 10.1103/PhysRevLett.100.066602 PACS numbers: 72.25.Ba, 72.25.Mk, 75.70.Cn, 75.75.+a

There has been a great deal of interest in studying the spin-dependent transport properties in ferromagnetic-nonmagnetic hybrid structures since the discovery of the giant magnetoresistance (GMR) effect [1]. The control of spin degree of freedom as well as its electronic charge for electronic applications is a key ingredient in spintronics, which may provide advantageous properties such as low-power switching and nonvolatile information storage [2]. Especially, electrical spin injection from ferromagnet (F) into nonmagnet (N) is an important technique for developing spintronics. When a spin-polarized current flows across the interface between F and N, the spin accumulation is induced in the vicinity of the F-N junction because of the sudden change in spin-dependent electrical conductivity [3,4]. Thus, one can induce the nonequilibrium magnetization in N. However, the accumulated spins in the N diminish through diffusion because of dispersed spin coherence. The length scale over which the traveling electron spin memorizes the initial direction is known as the spin-diffusion length. This is an important measure to realize the efficient spin injection. Therefore, understanding responsible physics on the spin relaxation process is essential to realize and optimize the spintronic devices.

According to Elliott [5] and Yafet [6], the intrinsic spin relaxation is initiated by the electron-phonon interaction with the spin-orbit coupling in clean samples [7,8]. Since the electron-phonon interaction is suppressed at low temperature, the spin-diffusion lengths in the clean samples become very long. On the other hand, in realistic materials such as polycrystalline metallic thin films, the influences of the extrinsic scatterings with impurities, defects, and boundaries are superimposed over the intrinsic spin relaxation process. The spin relaxation in such metals is governed by the electron-phonon scattering at high temperature, whereas the spin-diffusion length is determined by the extrinsic scattering at low temperature. The temperature dependence of the spin relaxation process can be investigated by the conduction electron spin resonance (CESR) technique [9,10]. However, it is difficult to determine precise temperature evolutions of the spin relaxation time from the CESR signal because of the intricate analysis based on the relation between the diffusion time and the spin relaxation time dependent on the experimental conditions such as the sample thickness and the temperature. Lateral F-N hybrid devices with electrical spin injection techniques also allow us to study spin relaxation processes in the normal metals. Johnson and Silsbee demonstrated that the temperature dependence of the spin relaxation time can be investigated by means of the electrical spin injection with Hanle effect in highly pure aluminum foil for the first time [4]. Fabian and Das Sarma showed that the spin relaxation time of the Al obtained in [4] is quantitatively reproduced by the calculation based on Elliot-Yafet theory [11]. Although the Hanle effect using the spin injection is applicable in a wider range of studies than the CESR, the analysis is valid only for the single interface, where the influence of the F voltage probe is neglected. F voltage probes with Ohmic contact is known to disturb the spatial distribution of the spin accumulation in N [12,13]. Therefore, in order to evaluate the spin relaxation time quantitatively using Hanle effect, the device should have the tunnel F contacts or very long separation distance between the injector and detector [4,14,15]. On the other hand, the spin-diffusion length in a normal metal can be simply evaluated by the separation distance dependence of the spin accumulation. Using a conventional lateral geometry with mesoscopic dimensions, spin-diffusion lengths for normal metals such as Cu, Al, and Ag have so far been evaluated at RT and liquid helium temperature [12,14–17]. However, there is no systematic study on the temperature evolution of the spin-diffusion length in such metals. Although the influence of the temperature is mainly explained by the electron-phonon scattering, other contributions on the temperature dependence may be found in the spin relaxation process. Here, we report the temperature dependence of the spin accumulation in lateral spin valves with Py/Cu Ohmic junctions.

Lateral spin-valve structure is prepared by means of the undercut resist mask and shadow evaporation technique. The bilayer resist consisting of 300-nm-thick 6% methyl-
methacrylate (MMA) /150-nm-thick 4% polymethyl methacrylate (PMMA) was prepared on a SiO$_2$ substrate. The electron-beam lithography with 50 kV acceleration voltage is performed to transfer the mask pattern into the top PMMA layer. The undercut structure schematically shown in Fig. 1(a) is obtained because of the backscattering of the electron beam and the high sensitivity of the MMA resist. First, the permalloy (Py) layer is formed by oblique evaporation in a vacuum of $10^{-9}$ Torr. After the Py deposition, the Cu is evaporated from the normal to the substrate in the different chamber with a base pressure of $2 \times 10^{-7}$ Torr. In order to reduce the magnetic impurity in the Cu film, the chamber for the Cu evaporation is separated from that for the Py evaporation. It should be noted that these two chambers are connected in vacuum. Figure 1(b) shows a scanning-electron-microscope (SEM) image of a prepared device. As in the image, the junction size between the Py and Cu is $140 \text{ nm} \times 220 \text{ nm}$. The separation distance between two Py wires is varied from 250 nm to 1650 nm. The junction size, which also affects the spin accumulation, is maintained in the same size for all devices. The thicknesses of the Py and Cu wires are 20 nm and 320 nm, respectively. The resistivities for Py and Cu wires are respectively 23.1 $\mu\Omega\text{ cm}$ and 2.35 $\mu\Omega\text{ cm}$ at RT and 17.1 $\mu\Omega\text{ cm}$ and 0.69 $\mu\Omega\text{ cm}$ at 5 K. To avoid the process-dependent dispersion of the characteristics such as resistivity and spin polarization, all the spin-valve devices were fabricated in the same batch. It should be noted that the evaporation machines used in the present study are different from those in our previous study [12]. The nonlocal spin-valve (NLSV) measurements were performed by using conventional current-bias lock-in technique with an amplitude of 0.15 mA. The external magnetic field $H$ is applied along the Py wire as shown in Fig. 1(b).

The field dependence of the NLSV signal for 250-nm separation measured at 20 K is shown in the inset of Fig. 2. Because of the different switching fields for two Py wires, the resistance states corresponding to the parallel (P) and antiparallel (AP) states are clearly observed. The magnitude of the resistance change $\Delta R_S$, which is defined by the resistance difference between the P and AP states, is as high as 2.9 m$\Omega$. We then measure the temperature dependence of the NLSV signal. As shown in Fig. 2, the spin signal monotonically decreases with temperature above 30 K. This reduction at $T > 30$ K is mainly due to the spin relaxation induced by the electron-phonon scattering in the Cu wire. However, the spin signal is found to decrease at $T < 30$ K as the temperature declines. Such a temperature dependence is observed in the lateral spin valves with different separation distances as shown in Fig. 2(b). We naively expected that the spin signal increase as the temperature is diminished and saturates at the low temperature because of the suppression of the electron-phonon scattering. However, such a simple prediction cannot explain the temperature dependence of the spin signals shown in Fig. 2. These indicate that other mechanisms for the reduction of the spin accumulation at low temperatures are required to explain these behaviors.

---

**FIG. 1 (color online).** (a) Schematic illustration of the fabrication procedure of a lateral spin valve by means of shadow evaporation technique. Py is evaporated at an oblique angle. The Cu is evaporated perpendicular to the substrate. (b) Scanning-electron-microscope image of the fabricated lateral spin valve consisting of two Py wires bridged by a Cu wire together with the probe configuration for the nonlocal spin-valve measurement. The location of Py wires are indicated by the dotted lines. The magnetic field direction is indicated by a solid arrow.

**FIG. 2.** (a) Temperature dependence of the spin signal in the Py/Cu lateral spin valve with the separation distance 250 nm. The inset shows the field dependence of the nonlocal spin-valve signal measured at 20 K. (b) Temperature dependences of the spin signals for different separation distances.
Now we evaluate the spin-diffusion length of the Cu wire from the spin signal measured as a function of separation distance $d$. The inset of Fig. 3(a) shows the spin signals versus $d$ at RT and 10 K. The spin signal decreases monotonically with the separation distance. By solving the spin-diffusion equation for the Py/Cu junction with the transparent interface, the dependence of the spin signal $\Delta R_S$ on the distance $d$ is given by \[ \Delta R_S = \frac{P_{Py}^2 R_{SPy}^2}{2R_{Py} \rho_{Py}} \exp\left(\frac{d}{\lambda_{Py}}\right) + R_{SCu} \sinh\left(\frac{d}{\lambda_{Cu}}\right). \] (1)

Here, $P_{Py}$ and $\lambda_{Cu}$ are the spin polarization for Py and the spin-diffusion length of Cu, respectively. $R_{SPy}$ and $R_{SCu}$ are the spin resistances for Py and for Cu, respectively. The spin resistance is defined as $2\rho \lambda/[1 - (1 - P^2)S]$, where $P$, $\rho$, $\lambda$, and $S$ are the spin polarization, the resistivity, the spin-diffusion length, and the effective cross section for the spin current, respectively. From the fitting curve, we obtain the spin-diffusion length for the Cu wire is 400 nm (at RT) and 1000 nm (at 10 K). The spin polarization and the spin-diffusion length for Py are 0.49 and 4.5 nm (at RT) and then 0.58 and 5 nm (at 10 K). The spin-diffusion length of the Py is in good agreement with previous reports \[12,14]. The spin polarization of the present Py wire is larger than that previously reported in the lateral devices (0.3–0.4) but is close to the value estimated from the current perpendicular to plane GMR experiments \[18]. The enhancement of the spin polarization in the present devices may be due to the in situ interface preparation.

The similar measurements were repeated every 10 K in order to evaluate the temperature dependence of the spin-diffusion length of the Cu wire. Figure 3(a) shows the temperature dependence of the spin-diffusion length of the Cu wire. Above 40 K, $\lambda_{Cu}$ monotonically decreases with increasing the temperature and seems to be inversely proportional to $p_{Cu}$ shown in Fig. 3(b). This means that the spin-diffusion length is proportional to the mean free path, supporting the presence of the electron-phonon interaction. However, surprisingly, $\lambda_{Cu}$ is found to take maximum at 40 K. Below 40 K, $\lambda_{Cu}$ decreases rapidly with the temperature. It should be noted that the spin-diffusion length and spin polarization of Py deduced from the experiments decrease monotonically with temperature. Using the parameters obtained from the fitting, the temperature dependence of the spin signal is quantitatively reproduced as shown in Fig. 3(c).

Thus, the reduction of the spin signal below 40 K is likely due to the reduction of $\lambda_{Cu}$. However, the mechanism of the reduction of $\lambda_{Cu}$ at low temperature is still an open question. The spin-diffusion length $\lambda$ is given by $\sqrt{D/\tau_{sd}}$, where $D$ and $\tau_{sd}$ are the diffusion constant and the spin relaxation time, respectively. As seen in Fig. 3(b), the resistivity of the Cu wire slightly decreases even below 40 K with the temperature, implying that the mean free path $l$ for the electrons in the Cu wire increases with decreasing the temperature. Since $D$ is proportional to the mean free path, the reduction of $\lambda_{Cu}$ is due to the reduction of $\tau_{sd}$. We believe that the surface oxidation is a possible origin for the reduction of $\tau_{sd}$. As Cu surfaces are known to be easily oxidized in air, the top and side surfaces of the Cu wire may be oxidized. The oxidized regions provide stronger spin-flip scatters than the inside of the Cu layer \[19]. Therefore, because of the increased mean free path at low temperatures, the effect of the scattering due to the oxidized Cu is more pronounced. This explains the reduction of the spin-diffusion length at low temperatures below 40 K. The ratio $\tau_{el}/\tau_{sd}$, where $\tau_{el}$ is the elastic scattering time, is known to represent the spin-orbit interaction strength \[5]. Using the relation $\tau_{el} = l/v_f$, where $v_f$ is the Fermi velocity, the ratio can be calculated as $2.74 \times 10^{-3}$ at 290 K and $4.64 \times 10^{-3}$ at 10 K. This means that the spin-orbit interaction becomes large at low temperatures, supporting that the reduction of $\tau_{sd}$ is due to the scattering of the oxidized layer.

To confirm this scenario, the dependence of the spin accumulation on the Cu thickness $l_{Cu}$ has been investigated. When the Cu thickness is decreased, the influence of the scatterings due to the oxidized Cu layer becomes large. Therefore, for thinner wires, the temperature $T_{M}$, where the spin signal takes a maximum value, becomes higher, since the mean free path is close to $l_{Cu}$ at higher temperatures. Figure 4(a) shows the temperature dependence of the spin signal for various $l_{Cu}$ from 30 nm to 320 nm. Here, the junction size between the Py/Cu and the thickness of the Py wires are the same as those in the above experiments. The distance between two Py wires is fixed to 250 nm. As

FIG. 3. (a) Temperature dependence of the spin-diffusion length of the Cu wire deduced from the distance dependence of the spin signal. The inset shows the spin signal as a function of the separation distance $d$ measured at 290 K and 10 K. The solid and open dots are the experimental data at 10 K and 290 K, respectively. The solid and dotted lines are calculated evolutions of the spin signal using Eq. (1) at 10 K and RT, respectively. (b) Temperature dependence of the resistivity of the Cu wire. (c) Calculated spin signal as a function of the temperature for $d = 250$ nm and $1650$ nm.
we expected, the increase in $T_M$ with decreasing $t_{Cu}$ is clearly seen as in Fig. 4(a). Figure 4(b) shows temperature $T_M$ as a function of $t_{Cu}$. $T_M$ gradually decreases with increasing $t_{Cu}$ while $t_{Cu} < 120$ nm. However, for the thick Cu wires with $t_{Cu} > 200$ nm, $T_M$ seems saturated around 35 K. This is because the main contribution of the boundary scattering shifts from the top to the side surfaces when $t_{Cu}$ exceeds 200 nm, which is the width of the Cu wire.

The spin signal also decreases with decreasing $t_{Cu}$, as shown in Fig. 4(a). Figure 4(c) shows the spin signal at 10 K as a function of $t_{Cu}$. According to Eq. (1) under the assumption that $\lambda_{Cu}$ and $\rho_{Cu}$ do not depend on $t_{Cu}$, the spin signal $\Delta R_s$ is roughly proportional to $t_{Cu}$. However, $\Delta R_s$ does not show a linear dependence on $t_{Cu}$. This can be understood as higher resistivity and shorter spin-diffusion length of the thinner Cu wire.

In conclusion, we have systematically studied the temperature dependence of the spin relaxation process in the Cu wires by means of nonlocal spin-valve measurement. The spin-diffusion length of the Cu wire decreases with increasing the temperature above 40 K because of the electron-phonon scattering. However, surprisingly, the spin-diffusion length is found to decrease below 40 K. This can be understood by the influence of the oxidized layer in the top and side surfaces of the Cu wire. The scenario based on the spin-dependent scattering with the oxidized layer well explains the experimentally obtained thickness dependence of the spin accumulation.