# Spin Detection and Injection using Ferromagnetic Metal and Semiconductor Hybrid Structure

T. Manago

Research Consortium for Synthetic Nano-Function Materials Project (SYNAF),
National Institute of Advanced Industrial Science and Technology,
AIST Tsukuba Central 4, 1-1-1 Higashi, Tsukuba, Ibaraki 305-8562, Japan
E-mail: t-manago@aist.go.jp

## Abstract

*We investigated spin-dependent transport properties from a viewpoint of spin detection and injection using a ferromagnetic metal / insulator ($Al_2O_3$)/ semiconductor tunnel junction with homogeneous and flat interfaces. For spin detection from the semiconductor, spin-polarized electrons were excited in the GaAs layer by circularly polarized light and injected into the permalloy layer. The energy dependence of the observed helicity asymmetry of the photo-induced current shows the absence of the spin-dependent tunneling. The result suggests importance of controlling the electron lifetime to obtain the spin-dependent tunneling. For spin injection into a semiconductor, we prepared Co/ $Al_2O_3$/ AlGaAs/ GaAs/ AlGaAs light emitting diode (LED) structure with ferromagnetic electrode. The electro-luminescence from the LED depends on the magnetization direction of the ferromagnetic electrode at room temperature. This fact shows that a spin-injection from the ferromagnetic metal to the semiconductor is achieved. The spin polarization due to the spin-injection current is estimated to be the order of 1 %.*

## 1. Introduction

Spin-dependent transport between ferromagnetic metals and semiconductors has been investigated intensively in recent years. This is based on an idea to use the spin of electrons in semiconductor electronic devices like a spin-FET proposed by Datta and Das [1]. Therefore it is one of the most important topics in "spintronics". However, there were two problems: whether one can electrically detect spin-polarized current in semiconductor using a ferromagnetic-metal electrode as a spin detector; and whether one can inject the spin-polarized electron from ferromagnetic metals into the semiconductors.

In earlier spin detection experiments, it was investigated using spin-polarized scanning tunneling microscopy using GaAs tip [2]. They studied spin-dependent transport properties of the photo-excited electrons from the semiconductor to the ferromagnet. Before this report, it had also been investigated using the ferromagnet/ insulator/ semiconductor tunnel junction for detail experiments [3], [4]. In GaAs, 50% spin-polarized electrons can be excited by circularly polarized light, in principle. When the ferromagnet has the spin polarization of 50 %, 25% of helicity asymmetry in the current is expected. However, A few percent of helicity asymmetry has been reported in the earlier experiments. This value is comparable to the magnetic circular dichroism (MCD). Therefore, it is delicate to distinguish the spin-dependent current from the MCD. We reported the absence of spin-dependent tunneling in the well defined tunnel junctions and discuss the problem [5].

Electrically spin injection was first realized using all-semiconducting devices [6]-[9]. These experiments were succeeded only at low temperature, since magnetic semiconductors does not work at room temperature. Therefore a lot of researchers make efforts to realize the room-temperature spin injection. Ferromagnetic metals are the likeliest candidates of spin-injecting sources. However, theoretical prediction showed that the limitation of the spin-injection efficiency from a metal into a semiconductor was less than 0.1 % due to a large conductance mismatch between them [10]. On the other hand, a tunneling process can overcome this problem, since it is not affected by the conductance mismatch [11]-[13]. The room-temperature spin injection was reported using a Fe/GaAs Schottky barrier contact [14], [15]. Since they used a reverse bias condition to inject electrons into GaAs, the tunnel contact was naturally made and the spin injection was done through the tunneling process across the Schottky barrier. The spin polarization is about 2 % at room temperature. Some groups tried to use tunnel barrier positively in order to obtain more spin injection efficiency. We reported room temperature spin injection using MIS structure [16]. Motsnyi *et al.* also reported in low temperature experiment using Hanle effect detection [17].

## 2. Experimental procedure

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The samples were grown by molecular beam epitaxy (MBE) with the III-V growth chamber, the metal growth chamber and the oxidation chamber without air exposure. The base pressure of these chambers was in the $10^{-10}$ Torr range. After the oxide removal of the substrate, atomically flat surface of p-GaAs(111)B (for spin detection samples) or light emitting diode (LED) structure consisted of $n^-$-$Al_{0.3}Ga_{0.7}As$/ SI-$Al_{0.3}Ga_{0.7}As$/ SI-GaAs/ SI-$Al_{0.3}Ga_{0.7}As$/ $p^+$-$Al_{0.3}Ga_{0.7}As$ /$p^+$-GaAs(001) substrate (for spin injection samples) were prepared. Subsequently, $Al_2O_3$ tunnel barrier with the thickness of 2 nm and the ferromagnetic electrodes with the thickness of 15 - 20 nm were grown. The film was covered with an Au capping layer. The homogeneity of our tunnel barriers on GaAs were checked by the thickness dependence of the junction resistivity [5] and a cross-sectional transmission electron microscopy image [16]. The logarithm of the junction resistivity is proportional to the thickness of the $Al_2O_3$ tunnel barrier, which is expected from the Wentzel-Kramers-Brillouin approximation. It indicates that the barrier is very homogeneous all over the sample. The junctions with a 60 μmϕ junction area were fabricated using the conventional photolithography, an argon ion etching, a sputter deposition of $SiO_2$ and metalization of electrode with an opening window for an optical access (48 μmϕ) to the junction. Back ohmic contacts were also formed by indium alloying at 200 °C. Schematic device structure is shown in Fig. 1 (a)

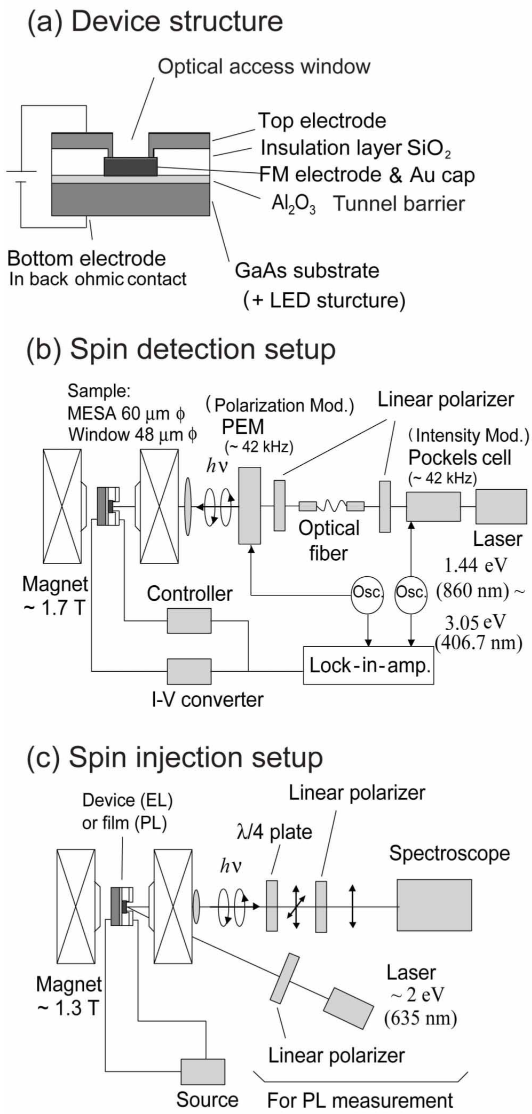


Figure 1. (a) Schematic junction structure of spin detection and spin injection devices. The junction area is 60 μmϕ and an opening window is 48 μmϕ for an optical access to the junction. (b) Schematic experimental setup for spin detection measurement. The photoexcited electrons modulated by Pockels cell and PEM were measured by a lock-in amplifier. (c) Schematic experimental setup for spin injection measurement. The spin-dependent electro-luminescence was detected by spectroscope with a quarter wave plate and an linear polarizer. Magnetic field is applied up to 1.3 T. For PL measurement, a laser (λ = 635 nm) and a subsequent linear polarizer were used.

For spin detection measurement (Fig. 1 (b)), the photo-excitation was performed by a mix-gas laser and a Ti-sapphire laser in the photon energy region of 1.44 eV (860 nm) - 3.05 eV (406.7 nm). The laser light was incident to the GaAs through the thin permalloy (Py) film normal to the plane. Intensity of the laser was modulated by the Pockels cell driving at about 42 kHz and the linear polarizer. The helicity was modulated by the photo-elastic modulator at the frequency of about 42 kHz. Both of the modulation depth was adjusted at maximum. The photocurrents by the light intensity modulation ($I_{IM}$) and by the circularly polarization modulation ($I_{PM}$) were measured by a lock-in amplifier. The bias voltage is applied to the top metal electrode with respect to the GaAs substrate. Magnetic field was applied up to 1.7 T normal to the plane, which was enough to saturate the magnetization of Py.

For spin injection measurement (Fig.1 (c)), the spin-dependent electroluminescence (EL) measurement was performed by a spectroscope (JASCO NRS-1000) with a quarter wave plate and an linear polarizer. It can analyze left and right circular polarization of the emitted light from the devices. The applied magnetic field (up to 1.3 T) and the direction of emitted light are perpendicular to the sample surface. We used a DC measurement and detected the polarization difference by reversing of the applied field. In order to estimate the MCD of the top ferromagnetic electrode, a photoluminescence (PL) measurement was also performed. The excitation photon energy was 2.0 eV. The laser light passes through a linear polarizer and the top ferromagnetic electrode. Even if the incident light is slightly polarized by passing the ferromagnetic electrode, the light with the photon energy of 2 eV hardly generate spin-polarized electrons [18]. Therefore we can ignore the MCD of the incident light and extract the MCD of the emitted light.

Both measurements were done at room temperature.

## 3. Spin detection

Permalloy is a suitable metal for the spin detection measurement because the spin polarization is high (about 50 %) [19] and the MCD vanishes at the vicinity of the photon-energy of the GaAs band-gap where the largest polarization is expected in the GaAs. The density of states of the Py calculated by the coherent potential approximation shows little energy dependence above the Fermi energy [20]. It is expected, therefore, that spin-dependent tunneling probability from the GaAs to the Py hardly shows energy dependence up to the upper edge of the minority band (about 1.4 eV from the Fermi energy). Thus in our experimental setup, we can detect the spin-dependent current exclusively. Fig. 2 (a) shows I-V characteristics of the tunnel junction. As is shown in Fig. 2 (b), the bias dependence of the intensity-modulation response ($I_{IM}$) shows that the photo-response increases in positive bias and very small response for negative bias is observed. Since the bias voltage is applied to the top electrode, the photo-exited electrons flow from GaAs substrate to the metal electrode in the case for positive bias.

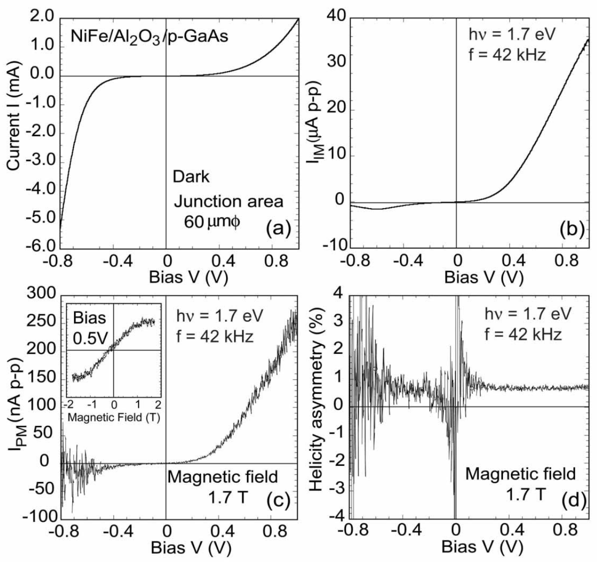


Figure 2. (a) *I-V* characteristics without illumination. (b) Bias dependence of the intensity modulation response of the photo-current. (c) Bias dependence of the polarization modulation response by the circular polarized light under the magnetic field (1.7 T) and magnetic field dependence of the response at $V$ = 0.5 V (inset). (d) Bias dependence of the helicity asymmetry ($I_{PM}$ / $I_{IM}$).

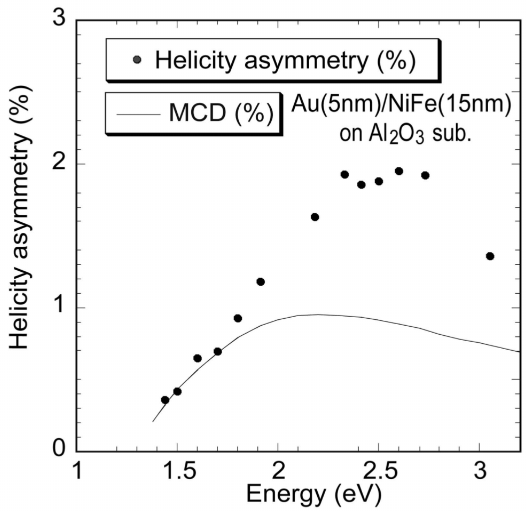


Figure 3. Energy dependence of the helicity asymmetry and the magnetic circular dichroism (MCD). The MCD sample is Au (5 nm)/NiFe (15 nm) on $Al_2O_3$ substrate. The thickness of Au and NiFe are the same as the transport measurement sample.

The polarization-modulation response ($I_{PM}$) in Fig. 2 (c) shows almost same behavior as the intensity-modulation response except the reduction of the intensity. As is shown in the inset, the sign of polarization-modulation response clearly depends on the direction of the magnetic field. The curve corresponds to the magnetization curve. We defined the spin-dependent signal (the helicity asymmetry) as the ratio of the $I_{PM}$ and $I_{IM}$, that is, $I_{PM}/I_{IM}$ (Fig. 2 (d)). The result seems that the spin-polarized current can be detected depending on the magnetization direction. Around bias voltage 0 V, it is divergent due to division by almost zero. There may be a possibility that the helicity asymmetry increases around 0 V, however, we cannot conclude it judging from the experimental accuracy. Whereas, the MCD of the magnetic electrode can generate the polarization-modulation current because the incident light through the top electrode causes the light intensity modulation by the different absorption rate for the right and the left circular polarized light. It should be noted that the helicity asymmetry is almost constant except the divergence. The tunnel magneto-resistance (TMR) device generally has the bias dependence of the TMR ratio that decreases with the bias voltage [21]. This decrease was considered to be due to the spin scattering in tunnel barrier. The observed bias independence implies that the helicity asymmetry comes from the MCD.

Fig. 3 shows the energy dependence of the helicity asymmetry and the MCD of NiFe film was also plotted. The MCD measured for a sample with the same Py thickness. It is well known that nearly 50% spin-polarization electrons are generated by the circularly polarized light at the band-gap energy of GaAs, and the spin-polarization decreases rapidly to 0 % around 2 eV [18]. In contrast, the observed helicity asymmetry of the photo-induced current gradually increases with the increase of photon energy. The MCD shows the qualitatively similar behavior. It is noted that the helicity asymmetry approaches zero as the photon energy approaches the band-gap energy of GaAs, which is the energy that the MCD becomes almost zero but the spin polarization in GaAs is maximum. These results clearly show that the helicity asymmetry is due to not the spin-dependent transport but the MCD effect.

Possible reasons why the spin-dependent tunneling was not observed in this experiment are (1) the spin-flip scattering at the permalloy/$Al_2O_3$ interface and/or (2) the spin -flip scattering at the GaAs/$Al_2O_3$ interface. Reason (1) can be excluded since the TMR effect is observed for the ferromagnetic metal/ $Al_2O_3$/ ferromagnetic metal junctions made by the same equipment on the GaAs substrates. Reason (2) is important because, in the *p*-type GaAs/$Al_2O_3$ junction, excited electrons diffuse toward the interface and stay there before tunneling. If the lifetime of those electrons is longer than the spin-relaxation time, one should not have the spin-dependent tunneling effect. Therefore, it is necessary to shorten the lifetime of the electron at the interface to obtain the spin-dependent tunneling. In addition, if there are electron traps and/or hole traps at the interface, photo-induced electrons will be trapped and then emitted from those traps. Emitted electrons have no spin-polarization if the lifetime of trapped electrons is very long. Concerning the spin-transport in the ferromagnet/ insulator/ semiconductor junction, Jansen *et al.* discussed the effect of the interface spin relaxation [22]. Sinohara *et al.* investigated the lifetime and the spin relaxation time of the micro-fabricated GaAs STM tips [23]. In our experiments, we found the modulation frequency dependence of optical responses of the photo-current. The optical response is larger for the lower frequency. This indicates the existence of the electron trap with very long lifetime of order of microseconds. This is one possible reason why we did not observe spin-dependent tunneling. To realize spin-detection device using GaAs and ferromagnetic metal, careful control of the electron-trap density seems to be essential.

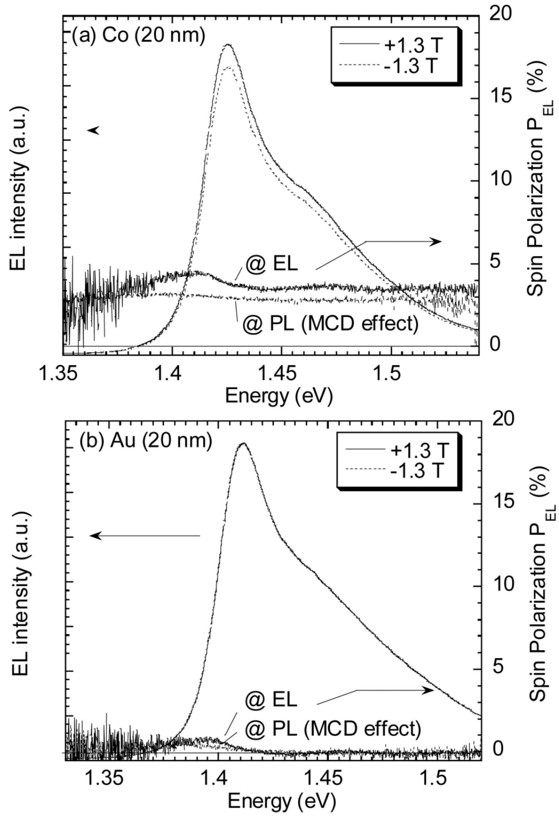


Figure 4. (a) The EL intensity of the LED with the Co electrode (left axis). Solid and dotted lines represent the spectra with the applied magnetic fields of +1.3 T and −1.3 T. The spin polarization *P* of EL and PL, $P_{EL}$ and $P_{EL}$, also plotted (right axis). The PL measurement represents the MCD effect of the top electrode. The difference between the $P_{EL}$ and the $P_{EL}$ corresponds to the spin-injection signal. All measurements were performed at room temperature. (b) The EL intensity of the LED with the Au electrode (left axis) and the spin polarization (right axis). It has no magnetic response. Slight deviation around the peak edge (≤ 1.41 eV) is an artifact that is from the lens of the apparatus.

## 4. Spin injection

Spin polarized electron injected from the ferromagnetic electrode into the GaAs well through tunnel barrier and the polarization of the emitted circular polarized light from the device was detected. The EL spectrum of the LED with a Co electrode at room temperature is shown in Fig. 4(a). The sample was biased at 2.8 V and the current is 22 mA. For comparison, the spectrum of the device without the ferromagnetic electrode (Au electrode) is shown in Fig. 4 (b). These spectra were observed at around 1.42 eV with a shoulder. The shape of the spectrums reflects the energy level of the quantum well states in the GaAs layer. The solid line and dotted line represent the spectrum with the applied magnetic field +1.3 T and −1.3 T, respectively. The spin-polarization is defined as $P = (I_+ - I_-)/(I_+ + I_-)$, where $I_+$ and $I_-$ are intensities in the case of + and − of the applied magnetic fields (the wave plate was fixed). Slight deviation at around the lower energy of the peak edge (≤ 1.41 eV) is an artifact reflecting the peak shape originated from a lens of the apparatus. Therefore we regard the polarization above 1.42 eV as an observed spin polarization. For the device with the Au electrode, it has no magnetic response. The *P* is almost zero above 1.42 eV. The result of $P \approx 0$ is unchanged in the case of rotating the

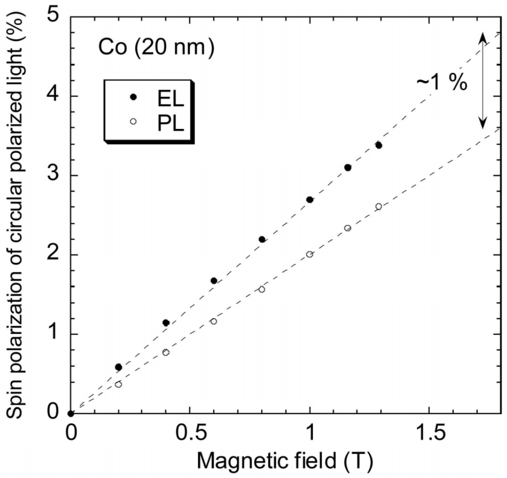


Figure 5. A field variation of the spin polarization of circular polarized light, the $P_{EL}$ and the $P_{EL}$. They increase in proportion to the magnetic field in the region. The difference between EL results and PL results is the spin polarization due to spin injection. It will reach about 1 % at 1.8 T, which can saturate the Co electrode. This means that the spin polarization of the electrons in GaAs reaches about 2 % under the saturation magnetic field.

wave plate 90-degrees. On the other hand, for the device with the Co electrode, the $P$ of the EL was estimated to be 3.5 %. Since the value includes the MCD of the top electrode, it is necessary that we extract effective spin polarization by spin-injection. The PL measurement is a useful tool for the purpose. The spin polarization of the PL $P_{PL}$ (%) of the same sample film was also plotted. The value is estimated to be 2.7 %, which represents the MCD effect of the top electrode. Therefore the effective spin-polarization ($P_{eff}$) is $P_{EL}$ (3.5 %) − $P_{PL}$ (2.7 %) = 0.8 %.

Fig. 5 is a field variation of the spin polarization. The magnetic easy axis of the samples lies in plane due to the shape anisotropy. The ferromagnetic electrode did not saturate magnetically at 1.3 T because the magnetic field was applied normal to the surface. The spin polarization increases in proportion to the magnetic field in the region. The difference between EL results and PL results means the spin polarization due to spin injection. It will reach about 1 % at 1.8 T, which can saturate the Co electrode. Taking into account the four-fold degeneracy of the valence band, it indicates the spin polarization in GaAs is $2P_{eff}$. Therefore the spin polarization of the electrons in GaAs is thought to reach about 2 % under the saturation magnetic field.

In our results, the spin polarization using the MIS structure was the same order of that of the Schottky barrier contact at room temperature. We think that in order to increase the spin injection efficiency, reducing the bias across the tunnel barrier seems to be necessary. As is reported in a magnetic tunnel junction, the tunnel magneto-resistance ratio (TMR) decreases with increasing sample bias [21]. The decrease is thought to be due to the spin scattering in the tunnel barrier [24] and the magnon scattering in the ferromagnetic electrode [25]. It was also reported that the bias dependence possibly originates from energy dependence of the spin-polarization around the Fermi level in the ferromagnetic electrodes [26]. To understand the bias dependence mechanism of the TMR is indispensable for higher spin-injection efficiency. And we also should take into account of the spin relaxation time in GaAs at room temperature.

## 5. Conclusion

We investigated spin-dependent transport properties from a viewpoint of spin detection and injection using a ferromagnetic metal/ insulator ($Al_2O_3$)/ semiconductor (GaAs or AlGaAs) tunnel junction with homogeneous and flat interfaces.

For spin detection from the semiconductor, we prepared the well regulated Py/ $Al_2O_3$/ p-type GaAs junctions and the spin-dependent transport of the photo-excited electrons was investigated carefully. The energy dependence of the observed helicity asymmetry of the photo-induced current shows the absence of the spin-dependent tunneling in the sample. The result suggests importance of controlling the electron lifetime to obtain the spin-dependent tunneling. In order to obtain spin-dependent current in the MIS structure, we need a large recombination velocity at the $Al_2O_3$/GaAs interface.

For spin injection into a semiconductor, we succeeded the spin injection from ferromagnetic metal into semiconductor using the Co/ $Al_2O_3$/ LED MIS structure. Spin-dependent electro-luminescence of the devices showed the spin-polarization in GaAs well is about 2 % at room temperature. The injection polarization was smaller than expected. It is probably due to the high sample bias across the tunnel barrier because in a magnetic tunnel junction, because TMR ratio generally decreases with increasing sample bias. It is necessary to operate at lower bias. We also should control the spin relaxation time in GaAs at room temperature.

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