Inversion of Spin Photocurrent due to Resonant Transmission

E. Wada,[1] Y. Shirahata,[1] T. Naito,[1] M. Itoh,[1] M. Yamaguchi,[2] and T. Taniyama[1,3,*]

[1]*Materials and Structures Laboratory, Tokyo Institute of Technology, 4259 Nagatsuta, Midori-ku, Yokohama 226-8503, Japan*

[2]*Graduate School of Engineering, Nagoya University, Furo-cho, Chikusa-ku, Nagoya 464-8603, Japan*

[3]*PRESTO, Japan Science and Technology Agency (JST), 4-1-8 Honcho Kawaguchi, Saitama 332-0012, Japan*

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We report on the inversion of spin-dependent photocurrent via interface localized states formed at the interface of an Fe/n-AlGaAs/GaAs quantum well heterostructure by means of an optical spin orientation technique. A careful adjustment of the excitation photon energy, which is determined by a separate analysis of electroluminescence spectra under a spin injection condition, enables us to explore the spin-dependent characteristics of photoelectron transmission from the quantum well into Fe. The bias dependence of the spin-dependent photocurrent shows clear spikelike features at the voltage which is compatible with the formation of the interface localized resonant states in the Schottky depletion layer.

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Electron spin transmission across a ferromagnetic metal (FM)-semiconductor (SC) interface is one of the most requisite issues to be explored for the development of spintronic devices. Quite a few studies of the electron spin transmission from a FM into a SC, and vice versa, have been carried out [1–15]. In order to shed light on the transmission mechanism, in particular, from a SC into a FM, a tremendous approach using an optical spin orientation method was proposed by Prins *et al.* [8], where spin-polarized photoelectrons are excited by the illumination of circular polarized light and the spin-dependent transmission across the interface was electrically detected: Circular polarized light can excite spin-polarized electrons efficiently in the SC according to the optical selection rules [11–13]. However, the mechanism of the electron spin transmission at the FM/n-GaAs interface is elusive and the understanding is still at a pioneering stage, presumably due to the following two underlying causes. Spin-polarized photoelectrons excited in GaAs near the Schottky interface predominantly drift into the GaAs bulk due to the built-in potential as shown in Fig. 1(a), while at the same time spin-unpolarized holes largely contribute to the charge transport across the interface. As a consequence, just a small number of spin-polarized electrons are able to transmit due to the diffusion process, giving rise to a slight detectable electrical signal of the spin-dependent photocurrent at the FM/n-GaAs interface. Furthermore, these previous studies have not been able to determine the accurate initial location as well as the energy of photoexcited electrons in GaAs; therefore, ambiguous information on the spin relaxation until the electrons reach the interface has to be taken into account rather speculatively.

In order to exclude the ambiguities stated above and probe the electron spin transmission more explicitly, we use in this study an Fe/n-AlGaAs/i-GaAs QW/p-AlGaAs heterostructure, where a significant built-in potential arising from the p-type SC/intrinsic SC/n-type SC junction is generated and a majority of photoexcited electrons now transmit towards the Fe layer [Fig. 1(b)]; thereby, the excited spin-polarized electrons can greatly contribute to the electron spin transmission signal at the Fe/n-AlGaAs interface. Moreover, prior measurement of electroluminescence (EL) from the quantum well (QW) under a spin injection condition from Fe into the QW can determine the electronic band information accurately, enabling us to specify the initial location and energy level of excited electrons in the QW. With this approach, here we investigate the electron spin transmission mechanism from a GaAs QW into Fe, and a remarkable inversion of the spin-dependent photocurrents which is likely associated with interface localized states formed at the Fe/n-AlGaAs interface is first detected.

The structures we use consist of the layer sequence of Au(10 nm)/Fe(5 nm)/n-$Al_{0.1}Ga_{0.9}As$(001) (75 nm)/undoped $Al_{0.1}Ga_{0.9}As$(001) setback layer (10 nm)/undoped GaAs(001) QW (20 nm)/undoped $Al_{0.3}Ga_{0.7}As$(001) (25 nm)/p-$Al_{0.3}Ga_{0.7}As$(001) (25 nm)/p-GaAs(001) buffer layer/p-GaAs(001) substrate. All the AlGaAs and GaAs layers were grown full epitaxially by molecular beam epitaxy with an amorphous

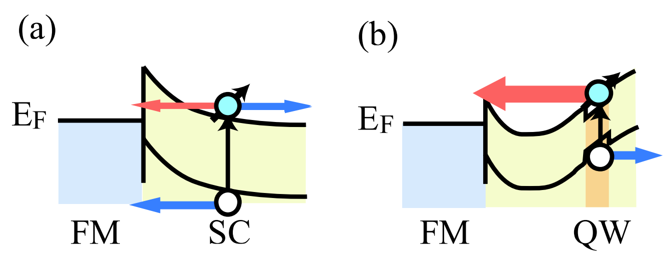

FIG. 1 (color online). Schematic diagram of photocurrent transmission at (a) an FM/n-SC interface and (b) an FM-SC QW (e.g., Fe/GaAs QW) interface. The red arrow denotes the spin-dependent photocurrent, while the blue arrows express spin-independent photocurrent contribution.

As capping layer on top of n-$Al_{0.1}Ga_{0.9}As(001)$. The QW structure was transferred to another Fe growth chamber and the As layer was decapped at 480 °C, followed by the growth of a 5-nm-thick Fe on top of n-$Al_{0.1}Ga_{0.9}As(001)$ at 30 °C. Prior to photoexcitation experiments, EL spectra under spin injection conditions from the Fe layer into the GaAs QW were measured for a bias voltage of −2.6 V at the interface to specify the excitation energy in QW, where the negative bias is defined so that the electrons transmit from Fe into GaAs. A magnetic field of 5 T was also applied perpendicular to the layer plane. Right-handed and left-handed circular polarized light (RCP and LCP, respectively) emission spectra were separately collected in a charge coupled device array equipped with a monochromator using a quarter wave plate and a polarizer. We then measured spin-dependent photocurrents excited by RCP and LCP light from an energy tunable continuous wave Ti:sapphire laser at the excitation energy obtained in the EL experiments, and the difference (ΔI) between the photocurrents was collected by means of a photoelastic modulator. All the photocurrent measurements were performed by using a lock-in technique. Spin-independent photocurrent (I_{ph}) was also collected in the same sample.

EL spectra for RCP and LCP components obtained at 10 K are shown in Fig. 2(a), consistent with previous reports [5,6]. To identify the origin of the EL peaks, we fit the spectra by the Voigt function which is the convolution of Lorentzian and Gaussian forms [16]; the decomposed four Voigt functions are given in the figure, and we assign the four decomposed peaks to features X, X_B, e-A^0, and X_B-LO. Since the energy of feature X is closest to the energy band gap of GaAs (1.519 eV) at 10 K and the spectra for RCP and LCP components split well, we attribute the feature to the recombination of free excitons in the QW. Bulk-related emission, i.e., free exciton recombination X_B, the conduction band to acceptor transition e-A^0 [5,17,18], and the LO phonon replica of X_B, X_B-LO [19], also occur in the GaAs buffer layer or substrate. Because the QW-related emission of X mainly contributes to the spin injection efficiency, we estimate the spin polarization of injected electrons by using $P_{spin} = 2P_{circ}$, where P_{spin} is the spin polarization of injected electrons and P_{circ} is the circular polarization of the emitted light [20]: No clear trace of the splitting of the heavy hole (HH) level and the light hole (LH) level is seen, and the splitting is estimated to be ~10 meV for our 20-nm-thick QW [1,2]. Figure 2(b) shows the magnetic field dependence of P_{spin} estimated from the integrated intensity of feature X and feature X_B. The P_{spin} originating from feature X is larger than that of feature X_B, ensuring that the feature X is due to the emission in the QW. It should also be noted that P_{spin} of feature X_B decreases above 2 T. A similar decrease was reported previously [3,4], and it could be attributed to the Zeeman thermalization in GaAs or AlGaAs. From the decomposed EL spectrum of the feature X which peaks at 1.508 eV, we decided to use the excitation energy of 1.503 eV for photoexcitation measurements, since spin-polarized electrons can be excited at around the minimum of the conduction band efficiently. The excitation energy of 1.666 eV was also used to generate hot electrons in the QW. These careful adjustments of the excitation energy now enable us to excite spin-polarized electrons in the conduction band of the QW. Also, the obtained spin polarization values up to 13% guarantee the good sample quality for photoexcited electron spin transmission experiments.

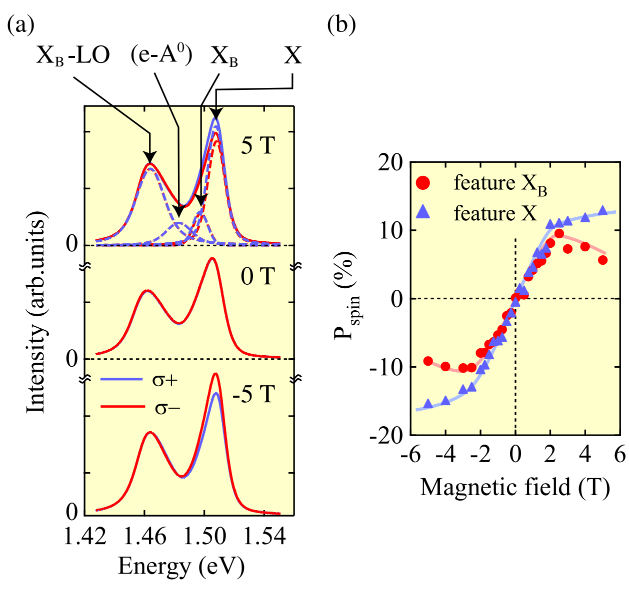

FIG. 2 (color online). EL spectra for LCP and RCP components obtained at 10 K. (a) The solid curves are the spectra of LCP($\sigma+$) and RCP($\sigma-$) emission at −5, 0, and 5 T. The dashed curves are the peaks decomposed by the four Voigt functions. (b) The magnetic field dependence of P_{spin} for the features X and X_B is shown.

The voltage dependences of I_{ph} and ΔI are measured as a function of bias voltage at the interface by using the excitation energy determined above. Since ΔI depends on the light intensity illuminated, we adopt $\Delta I_{SF} = \Delta I/2I_{ph}$ as a measure of spin-dependent transmission contribution. The voltage dependences of ΔI, I_{ph}, and ΔI_{SF} are shown in Fig. 3. The most significant features in the ΔI and ΔI_{SF} are spikelike dips at −0.058 and −0.179 V for the excitation at 1.503 and 1.666 eV, respectively. The features are totally spin-dependent signals since such a spikelike feature does not appear in the corresponding I_{ph}. Also, the spin-dependent signals are reversed as a negative magnetic field of −5 T is applied. In particular, we note that even the sign of ΔI_{SF} is inverted for the 1.666 eV excitation.

In order to explore the origin of the spikelike dips, the excitation process of spin-polarized electrons needs to be considered carefully, and here we propose the most probable excitation processes as depicted in Fig. 4(a). Since the excitation energy of 1.503 eV is adjusted to be the light emission energy arising from the recombination of free excitons, the photoexcitation at 1.503 eV should occur at the Γ point of the GaAs QW. For the excitation at 1.666 eV,

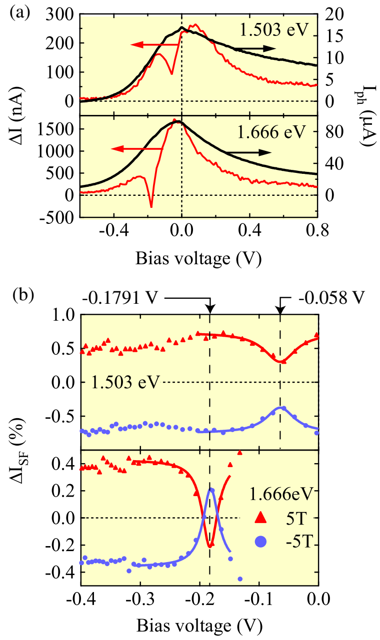

FIG. 3 (color online). (a) ΔI, I_{ph}, and (b) ΔI_{SF} under the excitation of 1.503 and 1.666 eV. The bias voltage of the spikelike dips are fitted by a Lorentzian function.

in contrast, hot electrons are likely generated so that the excited electrons have the k vector corresponding to the energy gap of 1.666 eV as shown in the figure. Therefore, in this case, the HH band and LH band split, and the excitation from HH predominantly occurs: The transition probability from the HH band is larger than that from the LH band [21], indicating that the photoelectrons are

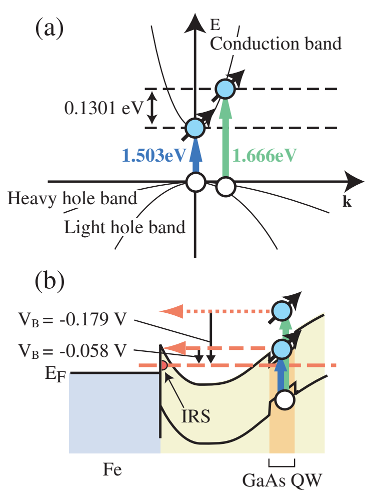

FIG. 4 (color online). Schematic diagram of (a) the excitation process in a QW and (b) spin-dependent transmission process of excited electrons at the Fe/GaAs QW interface. The blue and green arrows represent the excitation at 1.503 and 1.666 eV, respectively. The red arrows (dotted and dashed) express the transmission of spin-polarized electrons. $V_B = -0.179$ and -0.058 V denote the applied bias voltage. The dashed line is the energy level where the localized states are formed.

located 0.1301 eV above the conduction band minimum for the 1.666 eV excitation.

A possible spin-dependent transmission process from the QW into Fe is now illustrated in Fig. 4(b). At zero bias, photoelectrons generated in the QW at 1.503 eV excitation transmit across the Schottky barrier and show a spin-dependent transmission probability depending on the spin split density of states of Fe (dashed arrow). On the other hand, the spikelike decrease in ΔI_{SF} at a negative bias of -0.058 V clearly indicates that localized states are formed at the energy level 0.058 eV below the zero bias conduction band minimum of the GaAs QW (see the dashed line), where photoelectrons transmit into the Fe layer via the localized states. The shape of the spikelike dip is also well fitted by a Lorentzian function [see the solid line in Fig. 3(b)], indicating that the transmission is governed by the resonant tunneling via the localized states, based on the spin-dependent Breit-Wigner resonant formalism:

$$T_{R(L)}^{\uparrow(\downarrow)}(E) = \frac{4e^2}{h} \frac{\Gamma_F^{\uparrow(\downarrow)} \Gamma_A^{\uparrow(\downarrow)}}{(E - E_i)^2 + (\Gamma_F^{\uparrow(\downarrow)} + \Gamma_A^{\uparrow(\downarrow)})^2/4}, \quad (1)$$

where $T_{R(L)}^{\uparrow(\downarrow)}$, E_i, $\Gamma_F^{\uparrow(\downarrow)}/\hbar$, and $\Gamma_A^{\uparrow(\downarrow)}/\hbar$ are the transmission probability of spin up (down) electrons for the RCP (LCP) excitations, the energy level of a localized state, and the spin-dependent coupling between the localized state and Fe(AlGaAs) electrode, respectively [22]. $T_{R(L)}^{\uparrow(\downarrow)}$ is proportional to $\Gamma_A^{\uparrow(\downarrow)}/\Gamma_F^{\uparrow(\downarrow)}$ at the resonance ($E = E_i$), because $\Gamma_A^{\uparrow(\downarrow)} \ll \Gamma_F^{\uparrow(\downarrow)}$. Given $\Gamma_{F(A)}^{\uparrow(\downarrow)} \propto 1 \pm P_{F(A)}$, where $P_{F(A)}$ is the spin polarization of electrons in Fe (AlGaAs) [23], $T_R^{\uparrow(\downarrow)} \propto (1 \pm P_A)/(1 \pm P_F)$ and $T_L^{\uparrow(\downarrow)} \propto (1 \mp P_A)/(1 \pm P_F)$: The RCP (LCP) excitation provides the parallel (antiparallel) spin orientation between electrons in Fe and AlGaAs. Accordingly, ΔI_{SF} is expressed as

$$\Delta I_{\mathrm{SF}} \propto (T_R^{\uparrow} + T_R^{\downarrow}) - (T_L^{\uparrow} + T_L^{\downarrow}) = -\frac{4P_A P_F}{1 - P_F^2} < 0, \quad (2)$$

yielding a negative value of ΔI_{SF}. At off resonance ($E - E_i \gg \Gamma_F^{\uparrow(\downarrow)} + \Gamma_A^{\uparrow(\downarrow)}$), on the other hand, $T_{L(R)}^{\uparrow(\downarrow)}$ is proportional to $\Gamma_A^{\uparrow(\downarrow)}\Gamma_F^{\uparrow(\downarrow)}$, giving a positive $\Delta I_{\mathrm{SF}} \propto 4P_A P_F > 0$. Therefore, the signal inversion of ΔI_{SF} is understood in terms of the resonant tunneling via the localized states formed at the Fe/n-AlGaAs interface due to the asymmetry of the coupling between the localized states and Fe and AlGaAs, $\Gamma_A^{\uparrow(\downarrow)} \ll \Gamma_F^{\uparrow(\downarrow)}$.

Recently, interface resonant states for the minority spin channel at the Fe/GaAs interface were theoretically predicted [24,25]. Such interface resonant states are likely a possible origin of the resonant states here, and if that is the case, interface resonant states could significantly enhance the signal of the inversion since only the minority spin channel contributes to the resonant tunneling. The description of the resonant tunneling also gives a very comprehensive explanation for the bias dependence of ΔI_{SF} at 1.666 eV excitation. Since photoelectrons are excited at 0.13 eV higher than the conduction band minimum in this case, the difference between the bias voltages at which the dips appear in ΔI_{SF} for both excitations should be 0.13 eV, in good agreement with the value of 0.12 V obtained in this experiment. Also, recent theoretical studies showed that the interface states can change the sign of the spin polarization in spin extraction at a ferromagnetic metal-semiconductor interface [26] and spin-dependent density of states at the Fe/GaAs interface can help tunneling of minority electrons through the Schottky barrier, reducing the spin polarization significantly [27]. Both mechanisms could be responsible for the electron spin transmission across the Fe/AlGaAs interface we investigated.

One may consider that a small HH-LH splitting could contribute to the spikelike feature observed in the spin-dependent photocurrent measurements, because the ratio of the number of electrons excited from the HH and the LH, which are responsible for the electron transmission across the interface, should change with bias voltage and the net spin polarization decreases, accordingly. If that is the case, a monotonic reduction in ΔI_{SF} could appear; however, the spikelike feature and the inversion of the sign in ΔI_{SF} do not likely occur. Therefore, this effect cannot be a primary origin for the spikelike feature observed.

In conclusion, we have explored the spin-dependent transmission from a GaAs QW into Fe by using an optical spin orientation method. The use of well defined photon energy illuminated on the GaAs QW has enabled us to clearly detect spin-dependent photocurrent via interface localized states, where a significant spikelike dip of the spin-dependent photocurrent and even the inversion have been observed. From the results, the combination of electroluminescence measurement under the spin injection condition and photoexcitation experiments is found to be a very advantageous means to examine the electron spin transmission mechanism across the ferromagnet-semiconductor interfaces, which has not been detected in the previous approaches.

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*taniyama.t.aa@m.titech.ac.jp

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