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Bangladesh J. Sci. Ind. Res. **48(2)**, 131-136, 2013

BANGLADESH JOURNAL OF SCIENTIFIC AND INDUSTRIAL RESEARCH

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Optical orientation of electron spins and their relaxation mechanism in *n*-type GaAs

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Abstract

An investigation is performed to detect the optical orientation of electron spins and their relaxation in *n*-type GaAs. Electron spins are oriented by a circularly polarized light. When the photo-induced spin-polarized carriers are pushed by an external bias in the device, an optically spin-induced transverse voltage (V_S) is observed. The V_S resulting from spin current having two current contributions (spin drift and spin diffusion currents) is assumed to be proportional to the spin current or spin relaxation and is a measure of the net charge accumulation on the transverse edges of the sample device for the spin-polarized carriers. The observed spin polarization-dependent voltage signal is found to decrease exponentially with the applied electric field. It is also found that the signal decreases with increasing the lattice temperature. The results are discussed in comparison with a quantitative evaluation of the spin relaxation frequency of the photo-oriented electrons under drift in *n*-type zinc-blende semiconductors. The spin relaxation mechanism is also discussed.

Keywords: Spin transport; Spin orientation; Spin relaxation; Semiconductor

Introduction

Spintronics (Awschalom *et. al.*, 2002; Žutić, Fabian and Sarma, 2004; Miah, 2008a), or spin physics (Dyakonov and Khaetskii, 2008) in fundamental and a broader sense, having its root in magnetotransport or magnetoelectronics has been known since the discovery of the *anisotropic magnetoresistance effect* in 1857 by W. Thomas (Dyakonov and Khaetskii, 2008). It is a revolutionary new class of electronics based on the spin degree of freedom of the electron in addition to or in place of the charge and is an emerging research field in condensed matter physics. However, recent interest has been motivated by successful examples of metal-based spintronic devices, such as read heads for hard disc drives and magnetoresistive random access memory, based on ferromagnetic metals in which, as first suggested by Mott (1936), the electrical current is carried by independent majority and minority spin channels. These first metallic spintronic devices (passive components), discovered in 1988 (Baibich *et. al.*) [available as the first commercial product in 1994 (Daughton *et. al.*, 1994)], were sandwiched structures consisting of alternating ferromagnetic and nonmagnetic metal layers whose electric resistance depends strongly on the relative orientation of the magnetizations in the magnetic layers controlled by an external magnetic field.

The first semiconductor spintronic (active) device was suggested by Datta and Das (1990). They proposed an electronic analogue of an electro-optical modulator which was later termed as "*spin field effect transistor*" (spin-FET), in a two-dimensional (2D) electron gas contacted with two ferromagnetic electrodes (one as a source for the injection of spin polarized electrons and the other as an analyser or detector for electron-spin polarization). The Datta-Das spin-FET is a scheme to control the electronic spin with an electric field via the gate while it is travelling through the 2D transport channel. Since then, their proposal has been believed to be the most promising and led to an intense focus on realizing the semiconductor-based spintronic devices. However, due to the experimental difficulties in the efficient spin injection for a successful detection, the Datta-Das spin-FET is yet to be implemented in an efficient way. On the other hand, the optimization of the electron spin lifetimes and the detection of spin coherence are the major challenges in the field of quantum computation.

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Semiconductor spintronics was greatly motivated by the observations of long spin lifetime (τ_s) or spin diffusion length (δ_s) (δ_s=2 μm obtained by optical (Kikkawa and Awschalom, 1998) and δ_s=1.7 μm by electrical (Miah, 2008b) measurements) in semiconductors. However, one of the important requirements necessary in developing semiconductor spintronic devices is the detection of spin current (or spin relaxation) in a semiconductor (Miah, 2008a). For a reliable detection, the efficient transport of spin-polarized carriers through a semiconductor over reasonable distances within device dimensions is required. This is because if spin relaxes too fast, the distance travelled by an electron without losing its spin state will be too short to perform any practical purpose.

The detection of spin current in semiconductors has been obtained mostly by optical methods with varying degrees of success (Awschalom *et. al.*, 2002; Miah, 2009a; Miah, 2009b; Miah, 2009c; Ivchenko and Kiselev, 1992; Kato *et. al.*, 2004). However, an electrical means of detecting spin current or spin relaxation in semiconductors is desirable for possible device applications. An electrical detection of spin current in semiconductors has recently been reported (Dyakonov and Khaetskii, 2008, Lou *et. al.*, 2007; Miah, 2007; Miah and Gray, 2008). For example, Lou *et. al.* (2007) detected spin transport in a lateral ferromagnetic metal–GaAs semiconductor device in the presence of an external magnetic field at low temperatures. In a previous study (Miah, 2007), the spin current in GaAs was detected via the photo-induced anomalous Hall effect (AHE) (Miah, 2009d). Here an investigation to probe the optical orientation of electron spins and their relaxation in *n*-type GaAs has been reported.

Spin orientation

The generation of spin-polarized carriers, or spin polarization, usually means creating a nonequilibrium spin population. In optical excitation or pumping, the production of a carrier population with a net spin by the direct absorption of circularly polarized light ($\boldsymbol{\sigma}^{\pm} = (\hat{x} \pm i\hat{y})/\sqrt{2}$) is a consequence of the optical selection rules ($\triangle m_j = \pm 1$ for $\sigma^{\pm}$) for the heavy-hole (HH) and light-hole (LH) valence to conduction band (CB) transitions (Fig. 1). For optical pumping of bulk zinc-blende semiconductors, such as GaAs, with photon energy just above the energy bandgap (E_g), because of the selection rules governing optical transitions from HH, or LH, states to CB states, right circularly polarization ($\boldsymbol{\sigma}^{+} = (\hat{x} + i\hat{y})/\sqrt{2}$) generates a density of spin-down electrons ($n_{\downarrow}$) which is three times the density of spin-up electrons ($n_{\uparrow}$), and vice versa for left circularly polarized light ($\boldsymbol{\sigma}^{-} = (\hat{x} - i\hat{y})/\sqrt{2}$) (Pikus and Titkov, 1984). Hence, the initial electron spin polarization $P_e(0,0)$ generated by a σ^{+}, or a σ^{-}, beam in a zinc-blende bulk semiconductor, defined as $P_e(\vec{r},t) = (n_{\downarrow}(\vec{r},t) - n_{\uparrow}(\vec{r},t))/(n_{\downarrow}(\vec{r},t) + n_{\uparrow}(\vec{r},t))$, is ±0.5 (50%), provided that the photon energy is low enough to avoid exciting carriers from the split-off (SO) band. The polarization pumped with energies larger than the SO band to CB energy gap ($E_{SO\text{-}C} = E_g + \Delta_{SO}$) decreases because of exciting carriers from the SO band. Optical pumping with a $\sigma^{+}(\sigma^{-})$ light orients electron spins along the direction parallel (antiparallel) to the direction of the light propagation, i.e. along $-z(z)$ (Fig. 1). Optically excited hole spin relaxation is extremely fast (≤ 100 fs), therefore their polarization is effectively zero and need not be considered.

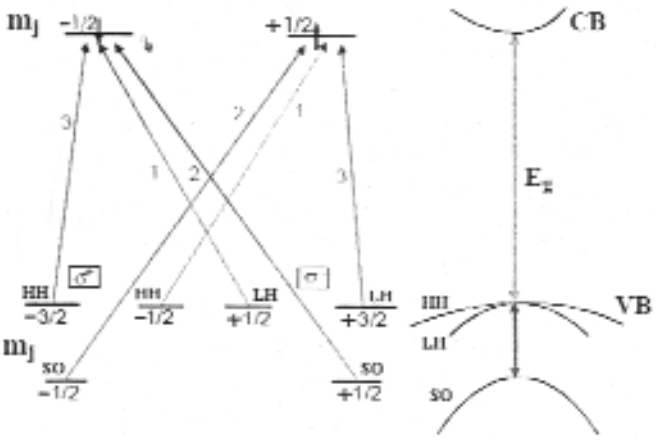

Fig. 1. Optical selection rules for the transitions from the heavy-hole (HH), light-hole (LH) and split-off (SO) valence bands (VB) to conduction band (CB). The allowed transitions for σ^{+} ($\triangle m_j$ = +1) and σ^{-} ($\triangle m_j$ = -1) are shown by the red and green lines respectively, where $\sigma^{+}(\sigma^{-})$ is for right(left) circularly polarized light. The numbers near the lines represent the relative transition probabilities. On the right, an *E* vs. *k* diagram of the energy bands of GaAs at *k*=0 shows the energy gap (E_g) and the spin-orbit splitting (Δ_{SO}) of the valence bands. The SO band to CB energy gap is $E_{SO\text{-}C} = E_g + \Delta_{SO}$. The degenerate states (energy levels) at *k*=0 are labelled on the left by their m_j quantum numbers. The small up (↑) and down arrows in CB indicate the electron's spin orientation.

Materials and methods

Investigated devices were fabricated on moderately (n =1×10^{16} cm^{-3}) silicon-doped (n-type) bulk GaAs.

Transparent Au/Ge/Pd contacts were deposited on the substrates. A gold wire was bonded to the centre of each of four contacts. The detail of the contact formation mechanism was reported elsewhere (Miah, 2009c). We optically excite (with excitation energy of ~ 4 mW) the sample (placed in a cryostat for low-temperature measurements) by circularly polarised picosecond pulses from a mode-locked Ti:sapphire laser with a repetition rate of 76 MHz. The polarization of the pulsed beam was modulated using a photo-elastic modulator (PEM) at lock-in reference frequency of 42 kHz. The excitation photon energy was tuned ($\lambda \approx 0.8$ μm) slightly above the band gap of GaAs. A NDW (neutral density wheel) was used to vary the optical power level. The laser beam was focused on to a ~90 μm (FWHM) spot at the surface of the sample with a lens. A lock-in amplifier was used for measuring the spin polarization-dependent voltage signal in the Hall geometry. For the low temperature measurements, the samples were housed in a temperature-regulated cryostat. A scheme of the experimental setup along with an illustration of the geometry of sample device is shown in Fig. 2. The round area at the centre ($x = 0$, $y = 0$, $z=0$) of the sample shows the light beam spot. The direction of the circularly polarized light beam (LB) propagation from the PEM is along $-z$. The electron spin-orientation is along $-z$ (z) for right (left) circularly polarized light σ^+ (σ^-).

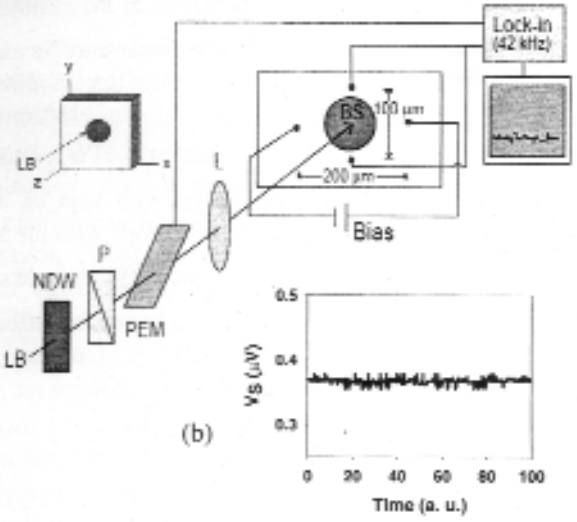

Fig. 2. A scheme of the experimental setup along with an illustration of the geometry of sample device (NDW: Neutral density wheel; P: Polarizer; PEM: Photo-elastic modulator; L: Lens; BS: Laser beam spot; and Look-in: Look-in amplifier). The red round area at the centre ($x = 0$, $y = 0$, $z=0$) of the sample shows the light beam spot. The direction of the circularly polarized light beam (LB) propagation from the PEM is along $-z$. The generation of spins is along $-z$ (z) for right (left) circularly polarized light σ^+ (σ^-). A typical scan for a sample for $E = 2.1$ kV cm^{-1} at 120 K is shown in (b)

Results and discussion

Experimental data

When the photo-induced spin-polarized carriers are dragged by an external bias in a sample, an optically spin-induced transverse voltage (V_S) is observed. The V_S is proportional to the spin current or spin relaxation and is a measure of the net charge accumulation on the transverse edges of the sample for the generated spin-polarized carriers. The origin of V_S is similar to the AHE, which has been known for a long time in ferromagnetic metals and in the presence of an external magnetic field.

Fig. 3 shows the electric field dependence of V_S for different lattice temperatures (T_L). A typical scan for a sample for $E = 2.1$ kV cm^{-1} at 120 K is given in Fig. 2b. The voltage signal decreases about exponentially with increasing the strength of E. This exponential dependence can be seen from the following expression for the spin polarization-dependent transverse electric field (Miah, 2008c) derived from the spin drift-diffusion model (Miah, 2008d).

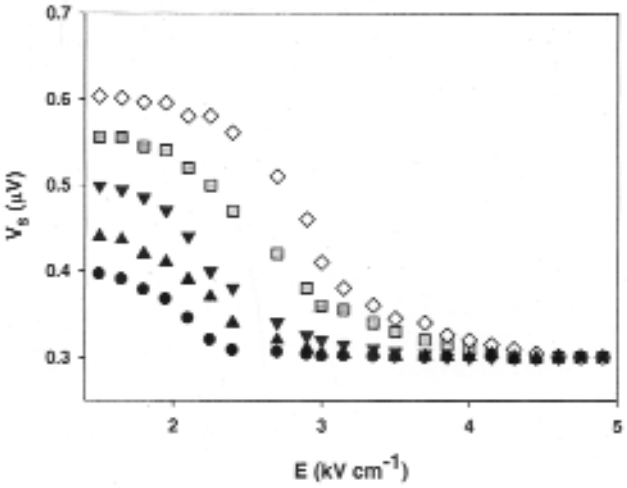

Fig. 3. Applied electric field dependence of the transverse signal for different crystal temperatures (circle: 300 K, triangle up: 120 K, triangle down: 40 K, square: 12 K and diamond: 4 K)

$$E_S = \sigma_S / (\sigma_D^2 - \sigma_S^2)(j + j_{dif})\, p(y) \qquad (1)$$

where $\zeta = \mu E/(2D_e)$ is the longitudinal electric-field parameter, D_e is the diffusion coefficient, μ is the drift mobility, $j = ne\mu E$ is the drift current, $j_{dif} = neD_e/\Lambda_S$ is the diffusion current, $p(y) = p_0 \exp(-y/\Lambda_S)$ is the density of spin polarization in the steady state and p_0 the

initial density of spin polarization (i.e. p at the point of spin generation, y=0), $\Lambda_S = 1/|-\zeta + \{\zeta^2 + 1/\delta_s^2\}^{1/2}|$ is the spin diffusion length in the presence of the applied electric field, σ_D is the Drude conductivity, σ_S is the anomalous Hall conductivity,. As can be seen from Eq. (1), the transverse field E_S has two current contributions (spin drift and spin diffusion currents) contributing to the total spin current. When E is very large ($\zeta \gg 1/\delta_s$), $1/\Lambda_S \approx 0$, and the diffusion current is zero, so that only the drift current contributes to E_S. This effect is illustrated in Fig. 4, where the reduced drift and diffusion currents are plotted as a function of the electric field and temperature, calculated for GaAs in the nondegenerate regime.

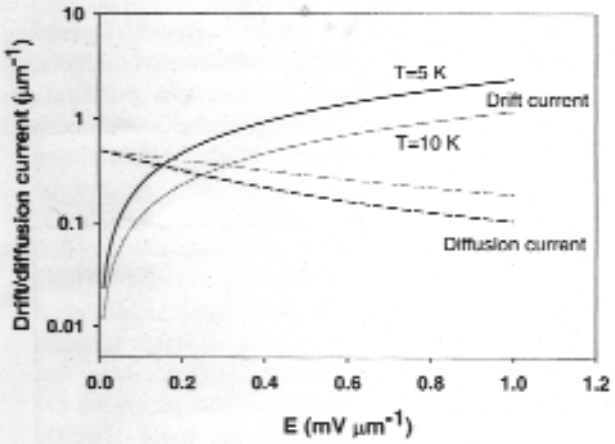

Fig. 4. Drift and diffusion currents (reduced by the quantity neD_e) as a function of the electric field and temperature in the nondegenerate electron statistical regime

As can be seen (Fig. 3), the spin polarization-dependent transverse signal increases with decreasing temperature, in consistence with the studies by Sanada *et. al.* (2002). These authors studied spin relaxation of photo-oriented electrons during transport in GaAs at low temperatures by the time-resolved photoluminescence polarization measurements. They showed that the polarization under drift increased with decreasing crystal temperature. The result is also consistent with the Faraday rotation measurements for the spin lifetime τ_s (Kikkawa and Awschalom, 1998).

As can be also seen, V_S for all temperatures decreases with E. The decay of V_S with E might be due to the enhanced electron spin relaxation at higher electric fields. The spin relaxation rate becomes considerably larger for higher fields. When the electrons were injected with a high E, a significant reduction in the spin relaxation is observed. Increasing E leads to larger charge and spin accumulations near sample boundaries, but polarization decreases because of shorter τ_s for larger E. The suppression of τ_s with increasing E implies that spin decay increases with E is consistent with other observations (Awschalom *et. al.*, 2002). From the variation of V_S it is revealed that the polarization suppresses exponentially.

The observed spin depolarization is considerably enhanced due to the enhanced spin relaxation as a result of increase of the electron temperature (T_e). As expected, electron spin at lower temperatures (compared to that at higher temperatures) disappears at higher fields. Our field-dependences agree with those of the transport experiment reported by Hägele *et. al.* (1998), where authors showed that the photo-oriented spins could travel without losing their initial spin orientation as long as E was below 1 kV cm^{-1} and the spin relaxation rate increased rapidly with E, and the polarization disappeared at ~3.5 kV cm^{-1}. It is noted that the photoelectron spins in their study were generated with a much higher optical power (10 mW).

Spin relaxation mechanism

The origin of field-dependent efficient electron spin relaxation in n-type GaAs is discussed based on the Dyakonov–Perel (DP) spin relaxation mechanism (Dyakonov and Perel, 1971). The DP mechanism is due to SO coupling in semiconductors lacking inversion symmetry. In III-V semiconductors, the degeneracy in the conduction band is lifted for $\vec{k} \neq 0$ due to the absence of inversion symmetry. Without inversion symmetry the momentum states of up-spin ($\uparrow$) and down-spin ($\downarrow$) spin electrons are not degenerate, i.e. $E_{\vec{k}\uparrow} \neq E_{\vec{k}\downarrow}$, where $E_{\vec{k}\uparrow}(E_{\vec{k}\downarrow})$ is the momentum-dependent electron energy with spin of $\uparrow(\downarrow)$. The resulting energy difference, for electrons with the same wave vector $\vec{k}$ but different spin states, plays the role of an intrinsic $\vec{k}$-dependent magnetic field, known as effective magnetic field (Pikus and Titkov, 1984),

$$\vec{h}(\vec{k}) = \alpha\hbar^2 / \sqrt{2em^* E_g}\,[k_x(k_y^2 - k_z^2)\hat{x} + c.p.] \qquad (2)$$

where $\hbar$ is the reduced Planck constant (Planck constant divided by 2π) and α is a dimensionless, material-specific parameter which gives the magnitude of the SO splitting and is approximately given by $\alpha \gg 4\eta(m^*/m_{cv})/\sqrt{3-\eta}$, where $\eta = \Delta/(E_g + \Delta)$, E_g is the energy bandgap, Δ is the SO splitting of the valence band, m^* is the electron's effective mass and m_{cv} is a constant close in magnitude to free electron mass m_0, induced by the presence of the Dresselhaus SO interaction in a zinc-blende structure (Dresselhaus, 1955), acting on the spin with its magnitude and orientation depending on $\vec{k}$, which results in spin

precession with Larmor frequency $\Omega_s(\vec{k})$ during the time between collisions, according to the relation $d\vec{S}/dt = \Omega_s(\vec{k}) \times \vec{S}$, where $\Omega_s(\vec{k}) = (e/m^*)\vec{h}(\vec{k})$ and $\vec{S}$ is electron spin polarization vector. The corresponding Hamiltonian term (DP Hamiltonian) due to spin-orbital splitting of the conduction band describing the precession of electrons in the conduction band is $H_{DP}(\vec{k}) = (\hbar/2)\vec{\sigma}\,\Omega_s(\vec{k})$, where $\vec{\sigma}$ is the vector of Pauli spin matrices.

The increased electron momentum at higher electric fields brings about a stronger $\vec{h}(\vec{k})$ and consequently, the electron precession frequency $\Omega_s(\vec{k})$ becomes higher. The effective magnetic field depends on the underlying material, on the geometry of the device, and on $\vec{k}$. Momentum-dependent spin procession described by the DP Hamiltonian mentioned above, together with momentum scattering characterized by momentum relaxation time $\tau_p(E_k)$ leads to DP spin relaxation. Since the magnitude and direction of $\vec{k}$ changes in an uncontrolled way due to electron scattering with the environment, this process contributes to spin relaxation, namely DP spin relaxation, given by (Pikus and Titkov, 1984)

$$f_{s,DP} = \gamma(\alpha/\hbar)^2 \tau_p E_k^3 / E_g, \qquad (3)$$

where $\tau_{s,DP}$ is the DP spin lifetimes and $f_{s,DP} = 1/\tau_{s,DP}$ is the DP spin relaxation frequency, $E_k = k_B T_e$ and γ is a dimensionless factor that ranges from 0.8 to 2.7 depending on the dominant momentum relaxation process. For example, for scattering by polar optical/ piezoelectric phonons $\gamma \approx 0.8$, while scattering by ionized impurities gives $\gamma \approx 1.5$, and scattering by acoustic phonons $\gamma \approx 2.7$ (Žutić, Fabian and Sarma, 2004).

The DP spin relaxation in a bulk zinc-blende structure occurs due to the spin precession about $\vec{h}(\vec{k})$ induced by the presence of the Dresselhaus SO interaction expressed in Eq. (5). During transport in the electric field, electrons are accelerated to higher velocities at higher fields, where T_e increases sharply due to the energy-independent nature of the dominant energy relaxation process via the longitudinal polar optical phonon scattering (Lundstrom, 1990). The resulting high T_e leads to enhanced DP spin relaxation because they have large kinetic energy between successive collisions; approximately the third-order power dependence of f_s on the electron temperature, i.e. $f_s \sim (k_B T_e)^3$.

In order to evaluate the DP spin relaxation quantitatively, we carry out a calculation of τ_s as a function of E. The optical-phonon scattering was included in the scattering in process ($\gamma \approx 0.8$), i.e. in the calculation of τ_p. The relation among drift-field, drift velocity, T_e and lattice temperature T_L was taken from (Lundstrom, 1990). Fig. 5 shows the calculated results. As can be seen, the spin relaxation frequency increases with increasing E. It is also seen that the spin relaxation frequency is rapid at higher fields and is almost infinite for fields higher than E=–3 kV cm^{-1}. This might be the result of an increase of the electron temperature at higher fields, and consequently, the higher DP spin relaxation frequency. The observed field-dependence agrees well with those of the experimental observations. The results are also consistent with the results of the simulation performed by others (Burry *et. al.*, 2003). They showed that for relatively low fields up to 1 kV cm^{-1}, a substantial amount of spin polarization is preserved for several microns at 300 K and the DP spin relaxation frequency increased rapidly for fields higher than ~1.5 kV cm^{-1}. It is thus concluded that DP mechanism is the dominant spin relaxation mechanism.

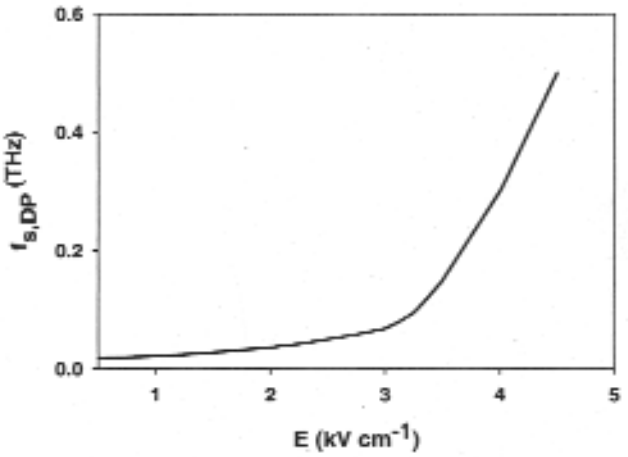

Fig. 5. Spin relaxation frequency as a function of applied electric field at T_L =4 K

Conclusion

On the basis of the AHE, optically oriented electron spins and their relaxation were detected electrically in a device fabricated on *n*-type GaAs. When the photo-induced spin-polarized carriers were dragged by an external bias in the device, an optically spin-induced transverse AHE voltage was observed. The observed AHE voltage signal was a measure of the spin current or the spin relaxation. It was found that the spin relaxation strongly depends on the applied electric field and lattice temperature. The spin polarization-dependent AHE voltage was found to decrease exponentially with the applied electric field, while it increased with decreasing the lattice temperature. A quantitative evaluation of the DP spin relaxation frequency

of the drifting polarized-electrons was performed. The experimental results were discussed based on the DP spin relaxation mechanism and were compared with those obtained in the calculations. A good agreement between them was obtained.

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Received: 15 May 2011; Revised: 05 September 2011; Accepted: 24 October 2011 .