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Room-temperature spin injection and spin loss across a GaNAs/GaAs interface

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Recently discovered effect of spin-filtering and spin amplification in GaNAs enables us to reliably obtain detailed information on the degree of spin loss during optical spin injection across a semiconductor heterointerface at room temperature. Spin polarization of electrons injected from GaAs into GaNAs is found to be less than half of what is generated in GaNAs by optical orientation. We show that the observed reduced spin injection efficiency is not only due to spin relaxation in GaAs, but more importantly due to spin loss across the interface due to structural inversion asymmetry and probably also interfacial point defects. © *2011 American Institute of Physics*. [doi:10.1063/1.3535615]

Efficient spin injection and reliable spin detection at room temperature (RT) are among the key challenges for future spintronics and spin-based quantum information technology.[1–3] In recent years, we have witnessed remarkable progresses on both electrical and optical spin injection and detection.[4–10] Unfortunately, the vast majority of earlier studies have been restricted to cryogenic temperatures. Demonstrations of spin injection/detection at RT, have started to emerge for ferromagnetic metal/semiconductor structures,[7–10] but generally with low efficiency. In principle, several mechanisms can be responsible for low spin injection efficiency. They include incomplete spin alignment within a spin aligner,[8] spin loss during interlayer spin transfer,[11,12] and low efficiency of spin detection.[13–16] The conductivity mismatch at a ferromagnetic metal-semiconductor interface has now been identified as a major cause for spin loss, which can be improved by inserting a tunneling barrier.[9] Structural defects such as stacking faults at a semiconductor-semiconductor interface were also shown to lead to strong spin scattering at low temperatures.[11] However, the extent of spin loss during spin injection across a semiconductor heterointerface at RT remains unknown.

A major difficulty in studies of spin loss at RT is a lack of reliable spin detector. Optical spin detectors based on polarized light emissions in semiconductors, successfully employed at low temperatures, have largely failed at RT due to accelerated electron spin relaxation with increasing temperature. Recently we demonstrated that spin-dependent recombination (SDR) via spin-polarized deep defects in GaNAs can selectively deplete conduction band (CB) electrons with the opposite spin orientation.[17] This so-called defect-engineered spin-filtering effect can be utilized not only to circumvent the limitation of spin relaxation imposed on spin detection efficiency, but also to amplify electron spin polarization. The aim of this work is, by exploiting this extraordinary ability of the GaNAs spin detector, to closely examine spin injection and spin loss across a GaAs/GaNAs interface at RT—the first case ever achieved for a semiconductor heterointerface.

Several GaAs/GaNAs structures with different N compositions, grown by molecular beam epitaxy (MBE) at temperatures T_g of 390–580 °C on a (001)-oriented GaAs substrate, were studied here. The growth started with a 2500 Å thick GaAs buffer, followed by either a 1000-Å GaNAs epilayer or seven-period GaAs/GaNAs (200/70 Å) multiple quantum-wells (QWs), and finally capped by a GaAs layer (200–1000 Å thick). These two different structures will be referred to as heterostructures (HSs) and QWs, respectively. In RT optical orientation experiments, photoexcitation at wavelengths of 750–980 nm was provided by a Ti-sapphire laser and was directed along the growth axis of the samples. Resulting photoluminescence (PL) signals were dispersed by a monochromator and detected by a Ge detector. Circular polarization of the excitation beam was generated by a $\frac{1}{4}$-wave plate.

For clarity, the principle of the SDR is schematically shown in Fig. 1(a). Under circularly polarized excitation (σ^+ or σ^-), spin blockade of carrier recombination via defects leads to: (1) higher spin polarization and (2) higher concentration of CB electrons, as compared with that under linearly polarized excitation (σ^x).[17–21] They in turn give rise to higher PL polarization and intensity, both providing a measure of CB electron spin polarization. Below we report on RT spin injection and loss across a GaAs/GaNAs interface, using the SDR ratio ($I^{\sigma^+}/I^{\sigma^X}$) as a means of spin detection. Here, I^{σ^+} and I^{σ^X} refer to PL intensity under σ^+, and σ^x excitation, respectively. The same conclusion can be drawn from spin detection by PL polarization.

In Figs. 1(b) and 1(c) we show representative RT PL spectra from the studied GaAs/GaNAs structures, under σ^+ and σ^x excitations. They arise from the band-to-band (BB)

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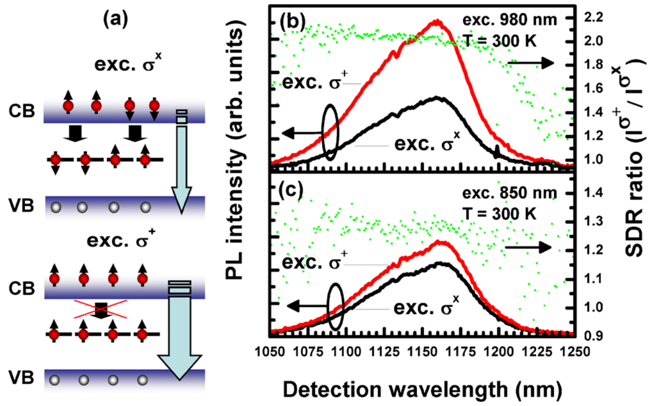

FIG. 1. (Color online) (a) Schematic illustration of the SDR effect on CB electron spin polarization and concentration, and thus PL polarization and intensity. [(b) and (c)] The representative RT BB PL spectra (solid lines) from the studied GaAs/GaNAs structures under σ^+ and σ^X excitation at 980 and 850 nm, together with the SDR ratio (dotted lines). The $\mathrm{GaAs/Ga_{0.974}N_{0.026}As}$ HS is taken here as an example.

transition in GaNAs.[17–21] With a fixed wavelength and constant power of excitation light, I^{σ^+} is consistently higher than I^{σ^X}, clearly manifesting the SDR effect. Additionally, the SDR ratio is noticeably lower upon spin generation at 850 nm (above the GaAs bandgap) than at 980 nm (below the GaAs bandgap but above the GaNAs bandgap), i.e., about 1.3 versus 2.1. This finding seems to indicate that the SDR effect is less effective when spin-polarized electrons are injected from the surrounding GaAs layers, as compared with spin generation within the GaNAs spin detector itself.

To firmly verify and also quantify the observed difference in the SDR effect, we carried out a detailed study of the SDR ratio as a function of the photoexcitation wavelength. A typical PL excitation (PLE) spectrum is shown in Fig. 2(a). It exhibits a distinct transition around the GaAs bandgap at around 870 nm that divides two regions of photogeneration, i.e., within GaNAs at the longer wavelengths and in GaAs at the shorter wavelengths. Apparently photogeneration of free carriers in GaAs is much more efficient than that within the GaNAs layer, by a factor of 4 judging from the PLE intensities. Therefore the PL from GaNAs in the former case must be predominantly induced from photogenerated carriers injected from GaAs and can be employed to study spin injection across the heterointerface. When the photoexcitation is below the GaAs bandgap, on the other hand, optical orientation within GaNAs is selectively studied.

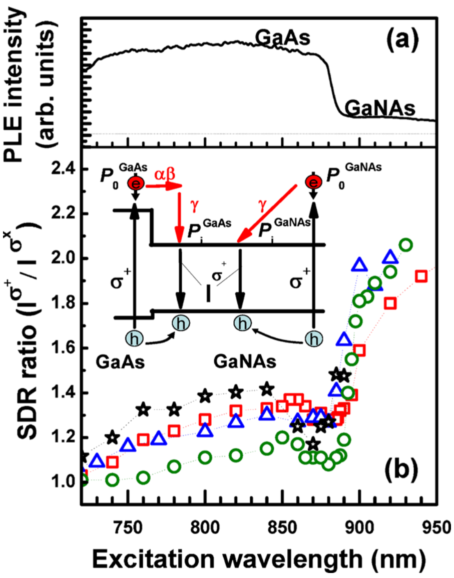

FIG. 2. (Color online) (a) Typical PLE spectrum obtained by monitoring the BB PL in GaNAs. (b) Values of the SDR ratio as a function of excitation wavelength from several GaAs/GaNAs HS and QWs: HS with [N]=2.6% and T_g=390 °C (squares), HS with [N]=1.3% and T_g=420 °C (circles), QWs with [N]=1.2% and T_g=420 °C (triangles), and QWs with [N] =1.1% and T_g=580 °C (stars). They were obtained by keeping a constant PL intensity at each excitation wavelength under σ^X excitation. The insert in (b) illustrates the spin generation, spin loss processes related to spin injection from GaAs and optical orientation within GaNAs.

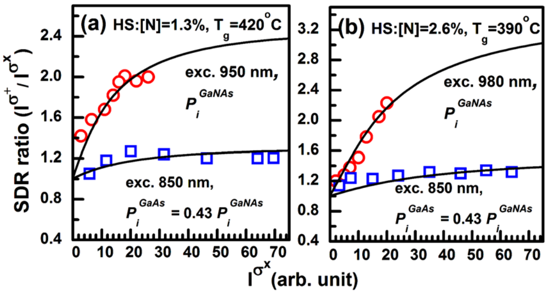

FIG. 3. (Color online) SDR ratio as a function of PL intensity under σ^X excitation, taken as examples from (a) the $\mathrm{GaAs/Ga_{0.987}N_{0.013}As}$ HS and (b) the $\mathrm{GaAs/Ga_{0.974}N_{0.026}As}$ HS. The circles and squares represent the data obtained under the excitation below and above the GaAs bandgap, respectively. The lines are the simulations from the rate equations analysis, yielding the values of the initial electron spin polarization given in each case.

In a given GaNAs sample, the efficiency of the spin-filtering process is determined by the following two factors:[17] (1) initial spin polarization and (2) the total number of the CB electrons before the spin-filtering takes effect. Here, we ensure that an equal number of the CB electrons are generated under both above and below GaAs excitation. This was done by adjusting excitation density at each excitation wavelength such that the intensity of the BB PL in GaNAs (scaled with photogenerated carrier density) remains the same under σ^x excitation. Now, the difference in the SDR ratio between above and below GaAs excitation is solely determined by the difference between the initial spin polarization induced by spin injection from GaAs (denoted by P_i^{GaAs}) and that created within GaNAs (denoted by P_i^{GaNAs}). The results from the various HS and QW structures are summarized in Fig. 2(b), which clearly show an abrupt and significant reduction in the SDR ratio once photoexcitation was undertaken in GaAs. This finding verifies that spin generation by spin injection from GaAs is less efficient than that through resonant excitation within GaNAs.

To confirm that this represents a general trend independent of carrier density, we have carried out a systematic investigation of the SDR ratio as a function of PL intensity in GaNAs under above and below GaAs excitation. The representative results are shown in Fig. 3, which confirm the trend revealed in Figs. 1 and 2 and provide compelling evidence for a weaker SDR ratio under the spin injection condition.

In order to estimate the extent of spin loss under spin injection, we have performed a detailed rate equation analysis of the results in Fig. 3 following the procedure given in Refs. 17–20. The analysis yields $P_i^{\mathrm{GaAs}}/P_i^{\mathrm{GaNAs}}=0.43$. In other words, the initial CB electron spin polarization in GaNAs generated through interlayer spin injection from GaAs is only 43% of that generated under resonant optical orien-

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tation within GaNAs, suggesting significant spin loss during the spin injection.

In principle, there could be several sources of spin loss for the observed reduced spin injection efficiency as illustrated by the inset in Fig. 2(b). The initial electron spin polarization in the light-emitting state of the GaNAs spin detector induced by spin injection and upon spin generation within GaNAs can be expressed by $P_i^{\mathrm{GaAs}}=P_o^{\mathrm{GaAs}}\alpha\beta\gamma$ and $P_i^{\mathrm{GaNAs}}=P_o^{\mathrm{GaNAs}}\gamma$, respectively. Here, P_o^{GaAs} (P_o^{GaNAs}) denotes the electron spin polarization generated in the instance of optical orientation in GaAs (GaNAs). α, β, and γ are the spin conservation factors associated with spin relaxation of electrons within GaAs before being injected to GaNAs, spin scattering across the GaAs/GaNAs interface and spin flips during energy relaxation of the injected hot electrons in GaNAs, respectively. Here, we assume similar spin loss during energy relaxation within GaNAs when the excitation photon energy was chosen just slightly above and below the GaAs bandgap. Values of P_o^{GaAs} and P_o^{GaNAs} are dictated by the selection rules of the electric dipole-dipole transitions, i.e., ~ 0.5 when both hh and lh VB states are involved as in our case.[22] Then, $P_i^{\mathrm{GaAs}}/P_i^{\mathrm{GaNAs}}=\alpha\beta$. The spin conservation rate during spin relaxation in GaAs can be determined by $\alpha = 1/(1+\tau^{\mathrm{GaAs}}/\tau_s^{\mathrm{GaAs}})$, where τ^{GaAs} and τ_s^{GaAs} are the total lifetime and spin relaxation time of the electrons in GaAs before being injected into GaNAs. τ^{GaAs} is governed by the spin injection time, known to be very short (<20–30 ps) from earlier studies.[17–20] τ_s^{GaAs} was measured by time-resolved PL in this work and is in the order of 70–100 ps. Based on these values, α can be estimated to be about 0.77. The spin conservation rate during spin injection across the GaAs/GaNAs interface can thus be deduced as $\beta=0.56$. In other words, spin loss by 44% is incurred during spin injection across the interface.

Below we shall briefly discuss possible mechanisms for the observed spin loss. A common cause for spin relaxation in a heterointerface stems from structural inversion asymmetry. The large CB discontinuity and an electric field due to interlayer charge transfer could lead to a large Rashba term, which promotes spin relaxation. Electron spin relaxation can also occur in the presence of defects at the interface. Earlier structural analyses showed that GaAs/GaNAs interfaces grown under optimal conditions are generally free of structural defects such as dislocations.[23] This excludes the possibility of structural defects as the source of spin loss.[11] Interfacial point defects have not been reported for a GaAs/GaNAs interface and are extremely difficult to detect and identify experimentally. Only until very recently was the first interfacial point defect at a semiconductor-semiconductor heterojunction reported—a $\mathrm{P_i}$ self-interstitial or $\mathrm{P_{Ga}}$ antisite at a GaP/GaNP interface.[24] Bearing in mind the similarity between GaAs/GaNAs and GaP/GaNP, the introduction of interfacial point defects during epitaxial growth is quite probable. They could act as efficient scattering centers for electron spins, leading to spin loss.

In summary, by employing the efficient GaNAs spin detector, reliable information on RT spin injection and spin loss across a semiconductor heterointerface is obtained for the first time. We have provided experimental evidence for significant spin loss (about 44%) during electron spin injection across the GaAs/GaNAs interface at RT. This is despite of the fact that the interface is free of structural defects. The observed spin loss is thus suggested to be promoted by the lack of structural inversion symmetry as well as possible point defects present at the interface.

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