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Electron spin control in dilute nitride semiconductors

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Abstract

We report on a study of spin-dependent recombination processes (SDR) for conduction band electrons on deep paramagnetic centers in a series of $GaAs_{1-y}N_y$ epilayers by time-resolved optical orientation experiments. We demonstrate that this dilute nitride compound can be used as an effective electron spin filter under a polarized optical excitation of appropriate intensity. This optimum intensity can moreover be controlled by adjusting the nitrogen composition in the layer.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Dilute nitride III–V semiconductors have been the subject of intensive research for the past decade thanks to the discovery of the dramatic change of the optical and electronic properties of certain III–V semiconductors (i.e. GaAs, InGaAs, GaP) induced by incorporating a small fraction (a few per cent) of nitrogen [1]. Due to their widely adjustable bandgap, these materials have been considered for the realization of devices for the telecommunication spectral windows ($\lambda = 1.3$ and 1.55 μm) [2, 3].

Very recent results [4–6] have shown that these dilute nitride compounds possess furthermore the remarkable property of a very long electron spin polarization even at room temperature, which could confer on them an important role in the spintronics field. Indeed, the creation, manipulation and storage of information under the form of electronic spin polarization in semiconductors represent the base of several propositions on future spintronic and quantum information devices [7, 8]. The creation of electronic spin polarization in semiconductors can be achieved by mainly two techniques: electrical spin injection through a magnetic layer [9, 10] and quasi-resonant optically oriented pumping [11]. The former shows a mostly limited efficiency at room temperature, while the latter requires, to achieve a strong degree of polarization, quasi-resonant excitation which can make the optical detection very difficult. Similar to bulk materials, once created, the spin-polarized carriers suffer from the unavoidable spin depolarization which in intrinsic layers can take place very rapidly (a few tens of picoseconds) [12]. The critical issue for the development of spin-related devices is still the conservation at room temperature of a sufficiently stable spin polarization, long enough to perform spin manipulations.

The notable electron spin property of dilute nitrides is due to a spin-dependent recombination (SDR) process, observed over 30 years ago in silicon [13] and (Al)GaAs [14, 15] and only very recently in GaAsN [4] with record high values. The results published to date on GaAsN, however, report data obtained only at room temperature. In this paper, we show that the SDR effect is effective from very low temperature ($T = 20$ K) and we demonstrate the use of the spin-dependent recombination mechanism of conduction band (CB) electrons in GaAsN as an efficient spin filter to maintain a strong electron spin polarization. The filter operates by effectively removing the spin-flipped electrons from the conduction band and trapping them non-radiatively on the centers. A temporally constant strong electron spin polarization, greater than the maximum value obtainable by

quasi-resonant optical pumping[5], can thus be created. We show that the SDR-related spin filtering effect is efficient from $T = 20$ to 300 K.

2. Samples and experimental set-up

The sample under study (sample I) consists of a 100 nm thick $GaAs_{1-y}N_y$ layer ($y = 0.021$), grown by molecular beam epitaxy on a (001) semi-insulating GaAs substrate and subject to a post-growth rapid thermal annealing. To explore the role of nitrogen content on the properties of electron spin filtering we have also studied a set of $GaAs_{1-y}N_y$ as-grown samples (samples II) with nitrogen content ranging from $y = 0.76\%$ to 2.6% and capped with 10 nm GaAs. We have investigated the spin properties in these nanostructures by time-resolved optical orientation experiments. The principle is to transfer the angular momentum of the exciting photons, using circularly polarized light, to the photogenerated electronic excitations. If the carriers do not lose their spin polarization during their lifetime, the photoluminescence (PL) will also be circularly polarized and will give information on both the symmetry of the carrier wavefunction and the spin relaxation time. The excitation source was a mode-locked Ti:Sa laser with a 1.5 ps pulse width and a repetition frequency of 80 MHz. The laser beam was focussed onto the sample to a 100 μm diameter spot and the PL intensity was dispersed by an imaging spectrometer. The temporal properties of the signal were recorded by an S1 photocathode streak camera with an overall time resolution of 8 ps. The excitation laser was either circularly (σ^+) or linearly (σ^X) polarized and the resulting PL circular polarization (P_c) is calculated as $P_c = (I^+ - I^-)/(I^+ + I^-)$. Here, I^+ and I^- are the PL intensity components co- and counter-polarized to the σ^+ excitation light. As the photogenerated hole spin relaxation time is very short [16] ($\approx$1 ps) P_c directly reflects the conduction band electron spin polarization degree.

3. Experimental results

In figure 1 we report the low temperature ($T = 20$ K) dynamics of the absolute value of P_c detected at the electron–light-hole transition, after a σ^+ polarized excitation for sample I. Due to the lattice mismatch, the growth of GaAsN on a GaAs substrate introduces a biaxial tensile strain, which lifts the degeneracy at the center of the Brillouin zone between heavy- (hh) and light-holes (lh) in the valence band, raising the lh band above the hh one as proved by optical orientation PL experiments [6]. At low temperature ($T = 20$ K) and under non-resonant σ^+ excitation (see footnote 5), the main contribution to the photoluminescence signal corresponds to the e–lh recombination as the thermal energy k_BT is much smaller than the separation $E_{lh} - E_{hh} \approx 24$ meV for $N \approx 2\%$. Because of the optical selection rules, the detected P_c is counter-polarized with respect to the laser [11].

[5] By non-resonant excitation we mean here an excitation energy greater than the electron–heavy-hole transition but smaller than the electron–split-off band. The maximum PL circular polarization achievable under this condition is $P_c = 50\%$.

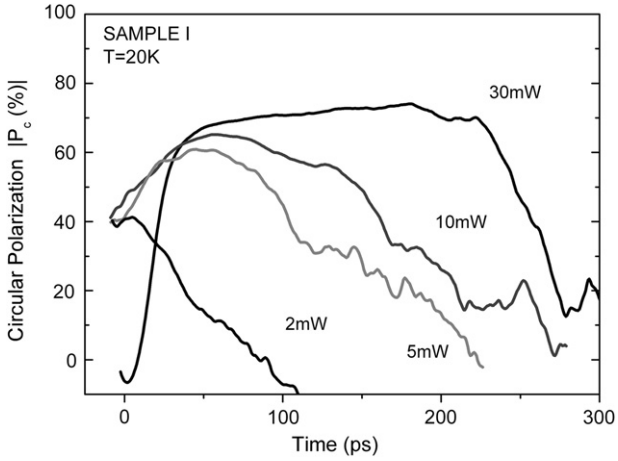

Figure 1. Time evolution of the absolute value of the PL circular polarization at $T = 20$ K for sample I, detected at the e–lh transition for different excitation powers. The laser excitation energy $E_{exc} = 1.392$ eV corresponds to the photogeneration of carriers directly in the tensile strained GaAsN epilayer. Similar results were obtained for an excitation energy above the GaAs bandgap.

At low excitation powers, $P_{exc} < 5$ mW, the P_c decay time is $\tau_s \approx 100$ ps. This value is in complete agreement with the measurement of the electron spin relaxation time in GaAs and other III–V compounds [17, 18] and is well explained in terms of the D'Yakonov–Perel mechanism [12]. For $P_{exc} > 5$ mW we can note the progressive appearance of a striking feature: the P_c decay time gradually lengthens as the excitation power is increased. For excitation powers $P_{exc} > 30$ mW, we observe the building up of a strong and stable degree of circular polarization $P_c^{MAX} \simeq 70\%$ which is greater than both its initial value $P_c(t = 0)$ and the maximum attainable value by quasi-non-resonant optical pumping (50%) according to the optical selection rules. This surprising result indicates that, in this compound, under sufficient excitation power, the 'apparent' electron spin lifetime τ_s^* can be much longer than the radiative lifetime ($\tau_R \approx 100$ ps). It is not possible to account for this power effect by simply considering a suppression of the D'Yakonov–Perel mechanism or the quenching of the classical spin relaxation mechanisms due to a strong localization of the electron wavefunction on N-related defects. The apparent increase of τ_s is instead due to a spin-dependent recombination process (SDR). The reader is referred to Lagarde *et al* and references therein for a comprehensive review [6]. The key point in SDR is the existence of deep paramagnetic centers which act as efficient non-radiative defects, created in GaAs by the introduction of small amounts of N. This process is schematically represented in figure 2(a). At each σ^+ laser pulse, the spin-polarized photogenerated electrons very rapidly dynamically spin-polarize the deep centers through a few non-radiative recombination cycles. For simplicity, we have assumed that the laser pulse created 100% CB electron spin polarization. Due to the Pauli principle, the CB electron capture is dependent on the relative spin orientation of the free and deep center resident electrons. After the capture, the deep centers can annihilate one of the electrons through a recombination with an unpolarized valence band

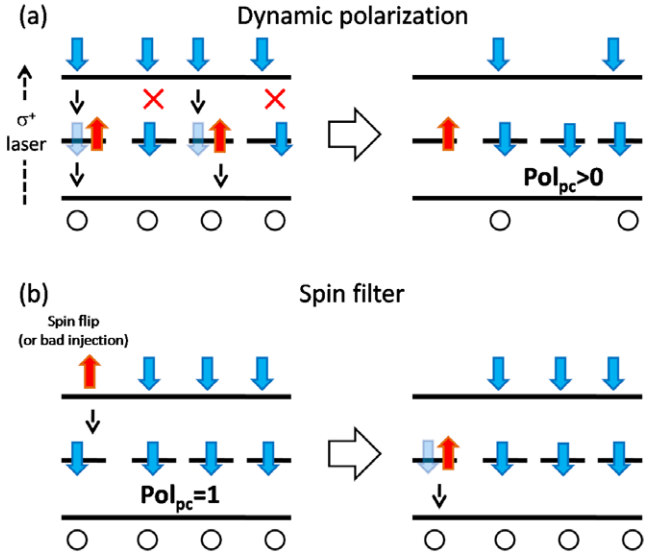

Figure 2. Sketch of the dynamical deep center polarization and spin filter process. (a) At $t = 0$, after a σ^+ laser pulse, the photogenerated spin-polarized conduction band electrons polarize the deep centers through a few cycles of non-radiative recombinations. For simplicity we have assumed that the laser pulse creates 100% electron spin polarization. (b) After being polarized, the deep centers act as a spin filter. If a CB electron suffered a spin-flip mechanism (or the electron spin polarization has to be increased), the polarized deep centers can readily capture the CB electrons of opposite spin. A high value of CB spin polarization is thus maintained. For illustration purposes we have assumed that the deep centers are fully spin-polarized.

hole. Once polarized, figure 2(b), the centers can now act as an effective non-radiative center only for conduction band electrons of opposite spin, provided that the centers keep their spin orientation for a time ($\tau_{s,pc}$) superior to the radiative lifetime and CB spin relaxation time ($\tau_{s,pc} > \tau_{PL}, \tau_s$). This is the feature upon which relies the principle of spin filtering: a spin-flipped electron is immediately captured by the spin-polarized deep centers. A strong spin polarization of the CB electrons can therefore be maintained, only the total PL intensity being affected.

In order to observe a sizable spin filtering effect it is necessary that the density of the CB electrons relative to the deep paramagnetic centers satisfies $n_e/n_{pc} \geqslant 1$: it would be otherwise impossible to achieve full deep center spin polarization. The remaining unpolarized centers will act as spin-insensitive non-radiative traps for the CB electrons. The strong power dependence of the P_c reported in figure 1 is well interpreted in terms of the ratio n_e/n_{pc}. As long as $n_e/n_{pc} < 1$ ($P_{exc} < 5$ mW) we recover the 'real' electron spin relaxation time τ_s as measured in GaAs. As the photogenerated carrier density increases, the spin filter regime is progressively reached and the maximum P_c (theoretically 100%) is attained.

The presence of a strong power-dependent SDR effect in this sample is experimentally confirmed as shown in figure 3. Figure 3(a) presents the time-integrated PL spectra under circularly (σ^+) and linearly (σ^X) polarized excitation as a function of the emission wavelength together with the respective ratio ($R = I^+/I^X$, open circles). This very high value, $R \approx 450\%$, peaked at the PL emission line is, to the best of our knowledge, the highest ratio reported in the literature for any semiconductor. This ratio is significantly higher than the corresponding value $R \approx 180\%$ obtained at room temperature [6]. Figure 3(b) displays the maximum SDR ratios for different excitation powers showing that an optimum spin-dependent effect is reached for $P_{opt} = 30$ mW. This is consistent with the effect observed in figure 1. Considering a 100 μm excitation diameter spot, the GaAsN absorption length and its reflection coefficient, we can estimate P $_{opt}$ corresponding to the photogeneration of an electron density $\approx 10^{17}$ cm^{-3}. The region below 30 mW in figure 3(b) corresponds to a number of photogenerated electrons insufficient to spin-polarize all the deep centers ($n_e/n_{pc} < 1$). The slight decrease of the SDR ratio above P_{opt} simply reflects the condition of an excess of photogenerated CB electrons ($n_e/n_{pc} > 1$) which cannot be filtered by an insufficient density of paramagnetic traps.

The efficiency of the spin filter effect at room temperature under the optimum excitation power P_{opt} is shown in figure 4, where the P_c dynamics for sample I are presented for laser excitations with different degrees of circular polarization. A linearly polarized excitation creates no CB electron spin polarization as an equal number of spin-up and spin-down electrons are created. An increase of the linear polarization degree (and a corresponding decrease of the circular one) can thus simulate the initialization of the system in a state of a different value of spin polarization. After the first picoseconds a strong P_c is recovered, whatever the initial degree of the excitation polarization is: the SDR-driven spin filter is effective as long as the system conserves a minimum initial imbalance of the spin-up and spin-down electron population and can be switched on by controlling the excitation intensity (i.e. $P_{exc} > P_{opt}$). It is worth noting that the rise time which characterizes the increase of the circular polarization in the first tens of picoseconds cannot be simply related to the dynamical polarization of the deep paramagnetic centers. This rise time is linked to the deep centers' polarization through a nonlinear system of coupled dynamical equations. According to the model by Kalevich *et al* [19] the circular polarization rise time is given by the hole recombination time $\tau_h^{-1} = \gamma_h N_{\uparrow\downarrow}$, where $N_{\uparrow\downarrow}$ is the concentration of paramagnetic centers with two electrons (centers that have captured one CB electron). By decreasing the electron spin polarization we increase the CB electron trapping probability, thus increasing $N_{\uparrow\downarrow}$. The resulting effect is a decrease of the hole recombination time which controls the deep centers polarization build-up, leading to a faster photoluminescence polarization increase.

We can systematically confirm the relation between the SDR mechanism and the N-induced defect density by studying the power dependence of the SDR effect as a function of the nitrogen content. To this aim, we performed the same investigation as for sample I on a set of $GaAs_{1-y}N_y$ samples (samples II) with varying nitrogen concentrations ranging from $N = 0.76\%$ to 2.6%, all grown under exactly the same conditions. For each sample we have reported in figure 5 (squares) the power corresponding to the maximum SDR effect, P_{opt} (N), at room temperature. As expected, the data shows an excellent correlation between P_{opt} and the nitrogen

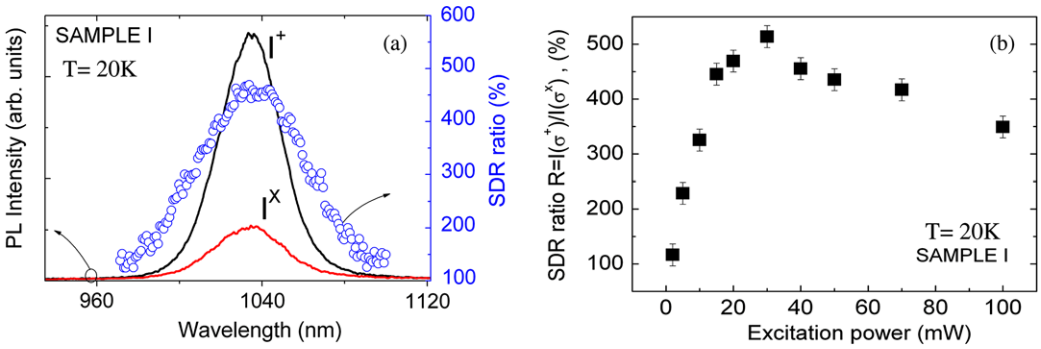

Figure 3. (a) Time-integrated PL intensity and the corresponding ratio $R = I^+/I^X$ under circular (σ^+) and linear (σ^X) excitations as a function of the emission wavelength for sample I. The excitation power and wavelength are $P_{\rm exc} = 40$ mW and $\lambda_{\rm exc} = 890$ nm ($E_{\rm exc} = 1.39$ eV), respectively, and $T = 20$ K. (b) Respective power dependence of the maximum SDR ratio under the same conditions.

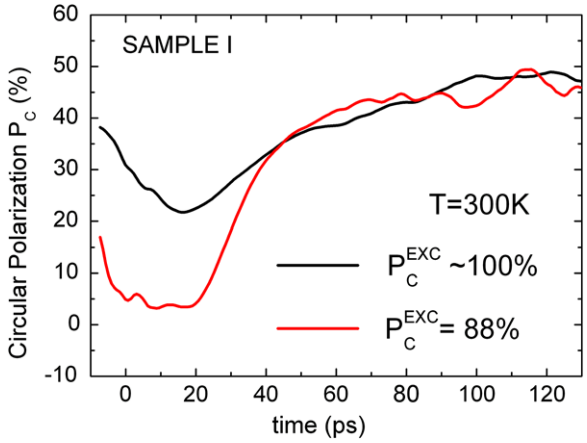

Figure 4. Room temperature time evolution of the PL circular polarization P_c for sample I under laser excitations with different degrees of circular polarization. $T = 300$ K, $E_{\rm exc} = 1.39$ eV.

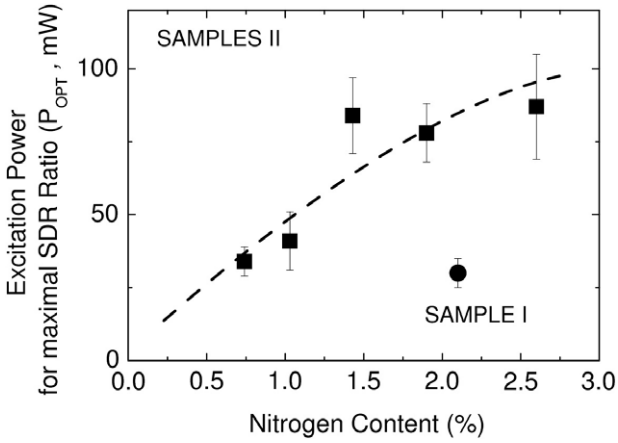

Figure 5. Excitation power to obtain the maximum SDR ratio ($P_{\rm opt}$) as a function of the nitrogen content in GaAsN for the set of samples II. $T = 300$ K, $E_{\rm exc} = 1.44$ eV. The circle represents the data for sample I (nominal nitrogen content $N = 2.1\%$) which underwent a post-growth rapid thermal annealing. The dashed line is a guide to the eye.

concentration, i.e. the defect density. It is not possible to extend this relation for nitrogen concentrations $y \geqslant 2.6\%$ as the PL intensity rapidly degrades, preventing the observation of the SDR effect. Figure 5 reports also the data (circle) corresponding to sample I. It appears that for sample I $P_{\rm opt}$ occurs at a much lower value than expected by comparison with the as-grown sample results. This is, however, not surprising as sample I has undergone a post-growth thermal annealing. The thermal treatment is, in fact, known to reduce sensibly the defect density in nitride compounds [20].

4. Conclusions

The introduction of a few per cent of N in GaAs leads to a radical modification of the electron spin dynamics, which is well explained in terms of a spin-dependent recombination through paramagnetic centers. This leads to record high values of the SDR ratio and opens up the possibility of using dilute nitride semiconductors as an effective spin filter for conduction band electrons even at room temperature. The spin filter is shown to be active as long as the system possesses an initial minimum spin polarization and can be activated by adjusting the excitation power. The systematic analysis of a set of samples with varying nitrogen concentrations confirms the dependence of the SDR mechanism on the defect density and provides a way to control its power dependence.

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