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Hole polarization and slow hole-spin relaxation in an *n*-doped quantum-well structure

Ph. Roussignol, P. Rolland, R. Ferreira, C. Delalande, and G. Bastard
Laboratoire de Physique de la Matière Condensée de l'Ecole Normale Supérieure, 24 rue Lhomond, 75005 Paris, France

A. Vinattieri, J. Martinez-Pastor,* L. Carraresi, and M. Colocci
Dipartimento di Fisica e Laboratorio Europeo di Spettroscopie Non-Lineari, Universitá di Firenze, Largo E. Fermi 2, 50125 Firenze, Italia

J. F. Palmier and B. Etienne
Centre de Bagneux, 196 avenue Henri Ravera, 92220 Bagneux, France
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We present a study of the dependence of the hole-spin relaxation on the electron density in an *n*-modulation-doped 75-Å $GaAs/Al_xGa_{1-x}As$ quantum well by using cw and time-resolved photoluminescence techniques, at low temperature. The electron concentration has been continuously varied from 10^{11} to 10^{12} cm^{-2}. A slow hole-spin relaxation time has been found (≈ 1 ns). A polarization decrease has also been observed when the in-plane wave vector of the photocreated holes increases. Calculations are presented which qualitatively support the latter experimental findings.

In recent years, a lot of work has been addressed to the dynamical behavior of the polarization induced by circularly polarized light in semiconductor heterostructures.[1] However, in spite of the large number of papers dealing with this subject, it still seems to be quite hard to extract a general behavior for the spin relaxation in these systems, due to the fact that the experimental results appear to be somehow sample dependent.[2] Moreover, the main results concern the dynamics of the polarization loss for *excitons*, for which the problem is clearly more complicated. Only a few studies have been addressed to polarization properties of independent carriers, *electrons* or *holes*, in quantum-well structures.

In bulk semiconductors, due to the fourfold degeneracy of the top of the valence band, holes relax their spin quasi-instantaneously. In quantum-well structures, where this degeneracy is lifted, a substantial change in the hole-spin relaxation process is expected.[3,4] Hole-spin relaxation times of the order of 4 ps[5] and 40 ps[6] have been reported.

In this paper we present an experimental study of cw and time-resolved photoluminescence (PL) on an *n*-modulation-doped 75-Å $GaAs/Ga_{1-x}Al_xAs$ quantum well embedded in a *p-i-n* diode. The p^+ substrate is followed by a 0.85-μm 8×4 $GaAs/Ga_{0.7}Al_{0.3}As$ superlattice, a 100-Å $Ga_{0.7}Al_{0.3}As$ barrier, and the 75-Å GaAs quantum well. The electrons transfer to the well through a 73-Å $Ga_{0.7}Al_{0.3}As$ spacer from a Si δ-doping plane. A 283-Å $Ga_{0.7}Al_{0.3}As$ layer and a 0.4-μm n^+ GaAs layer cap the structure. AuGeNi-Au contacts are taken both on the p^+ back side and the n^+ cap layer, leaving a 150-μm diameter window for optical measurements.

The electron density is estimated from measurement of the Stokes shift between the PL line and the first peak in the PL excitation (PLE) spectrum.[7] This Stokes shift varies from 60 (0 V bias) to 6 meV (13 V reverse bias), corresponding to an N_S variation from 10^{12} to 10^{11} cm^{-2}. A sizable current begins to flow through the structure at a voltage of 8 V and we could not achieve an electron concentration lower than 10^{11} cm^{-2}. For this range of electron concentration screening and occupancy effects are sufficient to prevent the existence of excitons. We will therefore, in a simple scheme, describe our results in terms of band-to-band transitions.

All measurements were done at $T=2$ or 4 K. Cw measurements have been performed by standard dye laser and lock-in techniques. For the time-resolved measurements the circularly polarized optical excitation (5 ps duration and 76 MHz repetition rate) was provided by a pyridine dye laser and the polarization-resolved photoluminescence was analyzed by a Hamamatsu synchroscan streak camera (20 ps effective resolution).

The cw measurements were performed at low excitation power: we estimate a photocreated carrier concentration of the order of 3.10^7 cm^{-2}. In Fig. 1(a) are reported typical PL and PLE results for a 6-V bias voltage. A Stokes shift of 33 meV ($N_S \approx 6\times10^{11}$ cm^{-2}) is observed between the PL peak and the first peak of the PLE spectrum corresponding to $E1^*H1^*$, the first band-to-band transition at the Fermi wave vector k_F. The second peak corresponds to the $E1^*L1^*$ transition. A small structure is also observable at higher energy (1.705 eV) corresponding to the band-to-band transition $E2H1$ at $k_\parallel=0$ (the quantum well is asymmetric due to the electric field).

A significant variation of the polarization of the PL line has been measured as a function of the excitation energy [Fig. 1(a)]; the polarization $P^{cw}=(I^+-I^-)/(I^++I^-)$ is maximum when exciting in the vicinity of the $E1^*H1^*$ transition and decreases steeply as soon as the excitation energy approaches the $E1^*L1^*$ transition. Moreover P^{cw} exhibits a secondary maximum for an excitation energy corresponding at the $E2H1$ transition. The dependence on the applied bias of $P_{E1^*H1^*}$, the polarization of the PL line when exciting at the $E1^*H1^*$ transition energy, and consequently its dependence on N_S, is depicted in Fig. 2(a). As N_S increases from 10^{11} to 10^{12} cm^{-2}, $P_{E1^*H1^*}$ decreases quasilinearly from 65 to 10%; in the same conditions P_{E2H1}, the polarization when exciting the quantum-

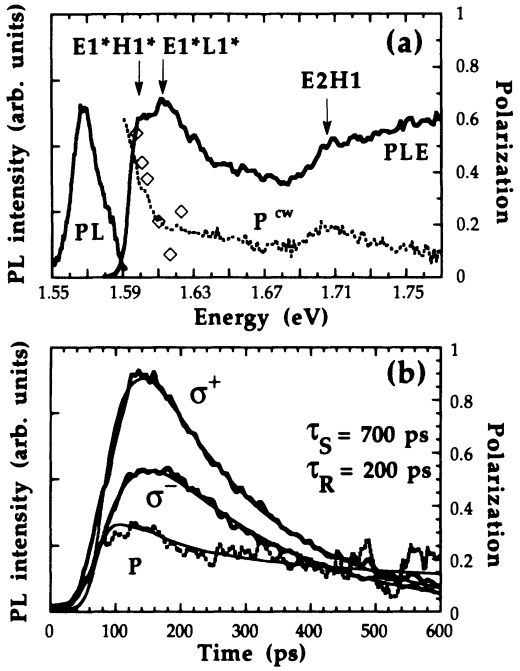

FIG. 1. (a) PL and PLE spectra of the 75-Å quantum well for a 6-V bias voltage (solid lines), P^{cw}, the cw polarization (dotted line), and $P(0)$, the time-resolved initial polarization (lozenges), as a function of the excitation energy are also reported. The relevant optical transitions are indicated by arrows. (b) Time-resolved right and left circularly polarized PL signals (solid bold lines) and polarization (dotted line) for the same bias voltage as (a). The solid lines correspond to theoretical fits with $\tau_R = 200$ ps and $\tau_S = 700$ ps.

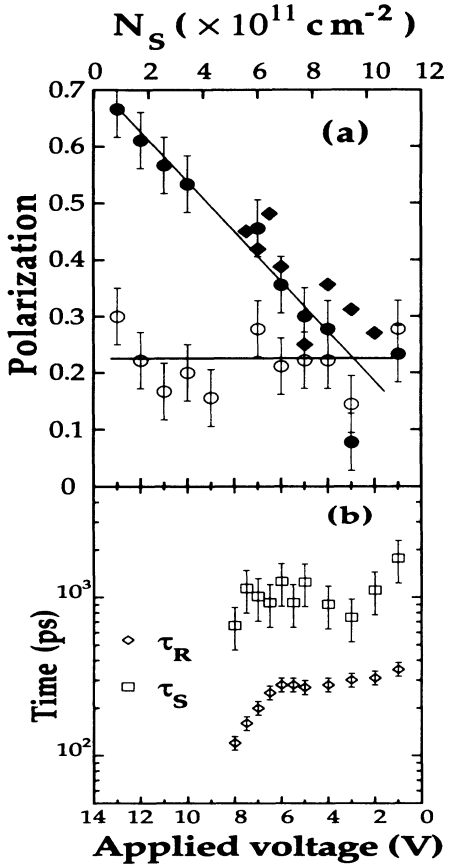

FIG. 2. (a) cw polarizations $P_{E1^*H1^*}$ (●) and P_{E2H1} (○) for excitation energies corresponding to the $E1^*H1^*$ and $E2H1$ transition energies of the 75-Å quantum well as a function of the applied voltage (lower scale) or the electron density (upper scale). The initial polarization measured in time-resolved experiments is also reported (♦). (b) Recombination time τ_R (◇) and spin relaxation time τ_S (□) as a function of the applied bias.

well structure at the $E2H1$ transition energy, remains constant at about 20%.

Time-resolved PL measurements after circularly polarized excitation give access to three different quantities: the initial polarization $P(0)$ of the system at $t=0$, the spin relaxation time τ_S, and the PL decay time τ_R. A typical experimental result is reported in Fig. 1(b) for an applied voltage of 6 V. After a right circularly polarized excitation, the time dependences of the PL signals $I^+(t)$ and $I^-(t)$ are recorded rotating the quarterwave plate located before the detection system (analyzer, spectrometer, and streak camera). The time dependence of $[I^+(t)+I^-(t)]$ and $[I^+(t)-I^-(t)]$, which decay with the time constants τ_R and $(1/\tau_R+2/\tau_S)^{-1}$, respectively, are then calculated and fitted.

For a given bias, the dependence of $P(0)$ on the excitation energy [lozenges in Fig. 1(a)] is similar with the one observed for the polarization P^{cw} after cw excitation [Fig. 1(a), dotted line]. In the same range of excitation energy, no significant variation of the recombination time and spin relaxation time is observed. The variations of $P(0)$, τ_R, and τ_S with the applied voltage have been studied between 0 and 8 V bias. In fact, no significant measurement was possible when the Stokes shift was smaller than 20 meV ($V > 8$ V). For each bias, the quantum well was excited at its $E1^*H1^*$ transition. The dependence of $P(0)$ on the bias voltage is reported in Fig. 2(a) (♦); it has to be noted that $P(0)$ has the same order of magnitude and exhibits the same behavior than the polarization $P_{E1^*H1^*}$ measured under cw excitation. In Fig. 2(b) τ_R and τ_S are reported as a function of the applied voltage; while τ_S remains fairly constant and of the order of 1 ns, τ_R decreases from 300 to 100 ps as the bias goes from 0 to 8 V. This decrease of τ_R occurs at the same voltage as a decrease of the cw PL efficiency and coincides with the appearance of a reverse current in the *p-i-n* diode. This variation is likely related to an evolution of the nonradiative channels responsible for the carriers recombination.

Photoluminescence from our *n*-modulation doped quantum well originates from band-to-band recombination of photoexcited holes and electrons of the Fermi sea. Due to the presence of a two dimensional electron gas in the well, the minimum in-plane wave vector at which holes can be photocreated is $k_{\parallel}=k_F$. When the quantum-well structure is excited at its $E1^*H1^*$ transition, heavy holes are created at $k_{\parallel}=k_F$, without superimposed excitation of light holes. The application of a bias, modulating the electron concentration, and consequently k_F, allows to study the optical properties of heavy holes photocreated at

different in-plane wave vectors.

In a first approximation, a right-circularly polarized excitation at the $E1^*H1^*$ transition energy produces heavy holes and electrons with respective spins $\frac{3}{2}$ and $-\frac{1}{2}$. In our time-resolved measurements the population of photocreated electrons (estimated to 10^{10} cm^{-2}) is always at least 1 order of magnitude smaller than N_S, so that the spin relaxation of electrons is irrelevant and the polarization of the electron Fermi sea can be disregarded. In the cw experiments, even when the excess electron concentration is small with respect to the equilibrium one, a continuous pumping of the Fermi sea may occur.[8] In our cw experiments, the concentration of photocarriers per unit time, N_C, is of the order of 10^{17} cm^{2}s^{-1} and the doping concentration N_S is at minimum 10^{11} cm^{-2}. Any saturation effect due to a polarization of the Fermi sea can be neglected in our measurements, the product $\tau_S N_C$ ($\approx 10^8$ cm^{-2}) being much smaller than N_S. Moreover, it is to be noted that, as shown in Fig. 2(a), cw and time-resolved measurements performed with excitation powers differing by more than an order of magnitude give totally coherent results for $P(0)$ and $P_{E1^*H1^*}$. Thus, all our measurements are related to the hole properties.

In a general way, let us call $G(k_\parallel)$ the polarization rate of the photocreated holes. By thermalization towards the band-edge part of the polarization is lost. The initial polarization $P(0)$ can therefore be written as a product, $P(0)=\alpha G$. The cw polarization $P_{E1^*H1^*}$ and $P(0)$ are related by $P_{E1^*H1^*}=P(0)/(1+2\tau_R/\tau_S)$.

The large value of τ_S (≈ 1 ns) with respect to τ_R (≈ 100 ps) explains why $P_{E1^*H1^*}$ is not significantly different from $P(0)$ [Figs. 1(a) and 2(a)]. The decrease of $P(0)$ with increasing initial $k_\parallel$ could be related to the decrease of $G(k_\parallel)$. In Fig. 3(a) we have reported the values of $(\langle J_z^2\rangle)^{1/2}$ (J_z is the projection of the angular momentum along the growth axis) for the $H1$ and $L1$ valence subbands (each twice degenerate) of a 75-Å symmetric quantum well as a function of the in-plane wave vector $k_\parallel$. Calculations are performed in the framework of the envelope function approximation using the Luttinger Hamiltonian which is diagonalized on the basis of the first three hole sublevels.[9] Considering the first heavy-hole level $H1$, due to the mixing of the valence bands at $k_\parallel \neq 0$ $(\langle J_z^2\rangle)^{1/2}$ varies from $\frac{3}{2}$ to 1.25 for $k_\parallel$ varying from 0 to 0.025 Å^{-1} (k_F at $N_S=10^{12}$ cm^2). Consequently G, which equals 1 at vanishing wave vector, decreases with increasing $k_\parallel$, but this effect remains small. The loss of polarization at $k_\parallel=0.025$ Å^{-1} is of the order of 10%, and does not affect very much the initial polarization $P(0)$.

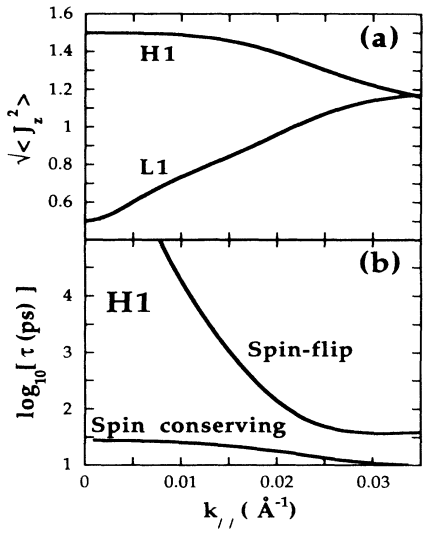

FIG. 3. (a) Square root of the mean value of J_z^2 for the first two valence subbands $H1$ and $L1$ of a 75-Å quantum well. (b) Dependence on the in-plane wave vector $k_\parallel$ of the emission times of acoustic phonons with (upper curve) and without (lower curve) spin flip for holes located on the first valence band $H1$.

More important is the term α which takes into account the fact that a hole population created at $k_\parallel \neq 0$ is always detected at $k_\parallel=0$. The polarization may be lost during the relaxation path towards the band edge, through emission of acoustic phonons or elastic scattering by ionized impurities or interface roughness. Let us call $H1\uparrow(k_\parallel)$ and $H1\downarrow(k_\parallel)$ the two energy degenerate $H1$ hole subbands. At a given time, a hole in a (say) $H1\uparrow(k_\parallel)$ state suffers scatterings which can relax its energy and/or its momentum. The final state can be either in the same up branch (spin-conserving scattering) or in the other one (spin-flip scattering). In Fig. 3(b) scattering times (without and with spin flip), calculated in the first Born approximation, are reported for processes assisted by acoustic phonons ($T=0$ K) described by the formalism of the deformation potential[10] as a function of initial $k_\parallel$. For $k_\parallel=0.035$ Å^{-1} the two times, τ^{ac} and τ_{sp}^{ac}, respectively, have the same order of magnitude and a nonnegligible probability of spin flip exists. As $k_\parallel$ decreases the emission time of an acoustic phonon without spin flip remains fairly constant; on the contrary τ_{sp}^{ac} increases dramatically with decreasing $k_\parallel$; near the band edge the spin-flip process becomes quite impossible for this process. The ionized impurity and interface defect assisted scattering times are also known to present the same behavior.[6] According to these calculations, the coefficient α, which results from a succession of such events, should decrease substantially as the wave vector, at which the holes are photocreated, increases.

The variation of α with $k_\parallel$ is very likely responsible for the experimental results depicted in Fig. 2(a). On the contrary, whatever the applied voltage is, when exciting the system at the $E2H1$ transition energy, holes are always mainly created at $k_\parallel=0$ and the polarization P_{E2H1} does not depend on the bias; the small value of 20% found for P_{E2H1} results from the fact that, as it can be seen on the PLE spectrum, the transition $E2H1$ contributes only partially to the absorption of the quantum-well structure; the other transitions involved in the absorption at 1.7 eV produce holes with large in-plane wave vectors and do not give rise to any polarization.

To summarize, we have studied by means of cw and time-resolved photoluminescence a *p-i-n* diode containing in its intrinsic part an *n*-modulation-doped GaAs/$Al_{0.3}Ga_{0.7}As$ quantum well. Varying the applied voltage we have investigated the spin polarization of holes as a

function of the electron density N_S. The polarization of the PL line corresponding to band-to-band recombination at $k_\parallel=0$ decreases with increasing N_S. According to calculations, the initial depolarization due to valence-band mixing is small. Thus, the observed loss of polarization before luminescence mainly occurs during the intraband relaxation path of the photocreated holes: the larger the Fermi edge, the smaller is α. As far as the dynamical properties of spin relaxation are concerned, for holes at $k_\parallel=0$, a very long spin relaxation time is observed (≈ 1 ns). This is in striking contrast with the bulk values ($\tau_0^{\text{bulk}} \approx 1$ ps). Our calculations do predict a considerable increase of $\tau_S(k_\parallel)$ with decreasing $k_\parallel$. However, a more complete theory is needed to render $\tau_S(k_\parallel=0)$ finite. We believe that it should include parity breaking terms (in z), neglected in the theory outlined above, such as the band-bending potential and/or the inversion asymmetry term due to the GaAs zinc-blende lattice. In fact, these small terms can be viewed as an effective $k_\parallel$ dependent magnetic field lying in the layer plane and coupling the $+\frac{3}{2}$ and $-\frac{3}{2}$ heavy-hole components. In close analogy with D'yakonov and Perels' work for electrons[8,11] a model for spin relaxation has to be undertaken for hole precessing around $B_\parallel$ and scattering on phonons or defects. Such a theory, clearly beyond the scope of this paper, would eventually explain long but finite $\tau_S(k_\parallel)$ for a thermalized distribution of holes near the band edge, in particular when the scatterers are numerous. In any case our experimental findings qualitatively supported by the theory clearly reveal the critical part played on the hole-spin relaxation by the lifting of the degeneracy between the heavy- and the light-hole subbands in quantum wells.

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*On leave from Departamento de Fisica Aplicada, Universitat de València, 46100 Burjassot, València, Spain.

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