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SPIN-DEPENDENT RECOMBINATION AND OPTICAL SPIN ORIENTATION IN SEMICONDUCTORS

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Spin-dependent recombination is observed in $Ga_{0.6}Al_{0.4}As$ at 77°K on the intensity of the donor–acceptor pairs photoluminescence. The lifetime is enhanced by a factor 2.3 when photocreated electrons and recombination centers are spin polarized by optical pumping with circularly polarized light. Optical orientation and spin-dependent recombination lead to a steady-state electronic spin polarization as large as 70%.

WE PRESENT in this paper an experiment which shows that spin-dependent recombination (SDR) in semiconductors[1] can be a very large effect. SDR originates from the Pauli principle which states that two electrons cannot have the same spin orientation in the same orbital state.[1,2] This leads to very different capture cross-sections of photocreated electrons by paramagnetic centers according to the relative spin orientations of the photocreated electrons and of the centers: when the spins of the recombining electron and of the center are parallel (triplet recombination[1,2]) the recombination probability is very weak as compared to the case where the spins are antiparallel (singlet recombination). In our experiments, spin oriented electrons and centers are created in zero magnetic field through optical pumping by circularly polarized light.[3–7] Due to SDR, the intensity of the photoluminescence light is then 2.3 times larger than when the excitation light is linearly polarized and creates no spin polarization. By measuring the degree of circular polarization of the luminescence which is proportional to the spin polarization of the recombining electrons, we show an additional feature of SDR: accumulation of electrons in one spin state giving a steady-state electronic polarization larger than expected from the absorption selection rules.

The experiment is performed at 77°K on a Ge-doped p-type $Ga_{0.6}Al_{0.4}As$ layer grown by liquid phase epitaxy on a GaAs substrate. The thickness of the layer is 50 μm and its energy gap is 2.03 eV at 77°K. The light at 1.92 eV from a krypton ion laser is focussed on the $Ga_{0.6}Al_{0.4}As$ sample. The polarization of the exciting light can be changed from linear to circular by rotating a quarter-wave plate. We observe the donor–acceptor pairs recombination of $Ga_{0.6}Al_{0.4}As$[8] at 1.90 eV by focussing the luminescence light on the entrance slit of a Spex 1700 spectrometer. We either measure the total intensity L_F of the fluorescence light or its circular polarized component $L_{F\pm}$ with $\sigma_\pm$ circular analysers. The degree of circular polarization of the fluorescence light $\rho = (L_{F+} - L_{F-})/(L_{F+} + L_{F-})$ is simply related to the steady-state electronic polarization $\rho = (n_+ - n_-)/(n_+ + n_-)$ (where $n_\pm$ is the density of recombining electrons with ± spin along the direction of propagation of the light) by the relation $\rho = -0.5\,p$.[5] We thus measure the electronic polarization by measuring the degree of polarization of the luminescence.

The steady-state polarization p depends on the polarization of the carriers at the instant of their creation and on the subsequent history of the electrons in the conduction band.[3] The initial polarization p_i is derived from symmetry arguments and is −0.5 in III–V compounds for band-to-band transitions under circularly polarized excitation and 0 under

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linearly polarized excitation.[5] In our case, since the energy of the incident photons ($h\nu = 1.92$ eV) is smaller than the band gap ($E_G = 2.03$ eV), the electrons are photoexcited from acceptor levels, probably germanium.[8] We assume that p_i for these transitions is the same as for band-to-band transitions because the wave functions of shallow acceptors are constructed from linear combinations of valence band wavefunctions.

The SDR photoluminescence ratio L_F(circ)/L_F(lin) is measured from the two curves of Fig. 1 and is found to be 2.3 ± 0.1 (L_F(circ) and L_F(lin) are the intensities of the luminescence under circularly and linearly polarized excitation), while the corresponding electronic polarizations are respectively p(circ) $= 0.70 \pm 0.01$ and p(lin) $= 0$. A very simple SDR model explains these results under the following assumptions:

(i) The lifetime is entirely controlled by recombination through centers of unknown origin which are paramagnetic before the capture of an electron.

(ii) The recombination process is completely forbidden when the spins of the recombining electron and of the center are parallel, that is the triplet capture cross-section is zero.

(iii) The paramagnetic centers which are unpolarized in the absence of light become polarized by their interactions with the polarized electrons. A possible mechanism is the following: a paramagnetic center can only capture an electron from the conduction band with a spin antiparallel to the one already present, whereas the electron released to the valence band is of either spin.

With these assumptions, we can write the following rate equations for the populations of electrons with + or − spin:

$$\frac{dn_\pm}{dt} = g_\pm - \frac{n_\pm}{\tau_\pm} - \frac{(n_\pm - n_\mp)}{2T_1} \qquad (1)$$

where T_1 is the spin relaxation time of photocreated electrons, $\tau_\pm$ and $g_\pm$ are the lifetimes and creation rates of electrons with ± spins. The recombination rates $\tau_\pm^{-1}$ are proportional to $N_\mp$, number of centers with spin *opposite* to that of the recombining electron:[1,2] $\tau_\pm^{-1} = \tau_0^{-1}\, 2N_\mp/(N_+ + N_-)$, τ_0 being the lifetime when there is no polarization . The creation rates are such that

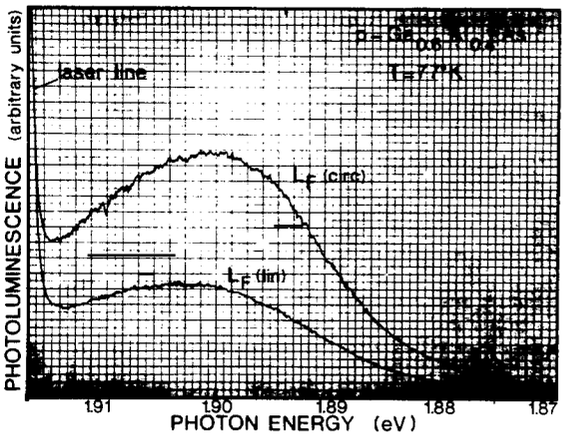

FIG. 1. Donor–acceptor pairs photoluminescence line of p-type $Ga_{0.6}Al_{0.4}As$ at 77° K. L_F(circ) and L_F(lin) are obtained under circularly or linearly polarized excitations with the same number of incident photons.

$$(g_+ - g_-)/(g_+ + g_-) = (g_+ - g_-)/g = p_i$$

Solution of these rate equations shows that the density $n = n_+ + n_-$ of photocreated electrons and their polarization p are:

$$n = g\tau_0 \left(1 + \frac{T_1}{T_1 \pm \tau_0} Pp_i\right)\left(1 - \frac{T_1}{T_1 \pm \tau_0} P^2\right)^{-1} \qquad (2)$$

$$p = \frac{T_1}{T_1 + \tau_0}(p_i + P)\left(1 + \frac{T_1}{T_1 + \tau_0} Pp_i\right)^{-1} \qquad (3)$$

where $P = (N_+ - N_-)/(N_+ + N_-)$ is the polarization of the recombining centers. The SDR photoluminescence ratio L_F(circ)/L_F(lin) is given by:

$$\frac{L_F(\text{circ})}{L_F(\text{lin})} = \frac{n(\text{circ})}{n(\text{lin})} =$$

$$\left(1 + \frac{T_1}{T_1 + \tau_0} Pp_i\right)\left(1 - \frac{T_1}{T_1 + \tau_0} P^2\right)^{-1} = 2.3 \qquad (4)$$

The value of $T_1/(T_1 + \tau_0)$ is 0.8 and is obtained from the decrease of ρ when a transverse magnetic field is applied as will be shown below. Inserting this value in the expressions of p and L_F(circ)/L_F(lin), we find that the results are consistently explained if one assumes that the polarization of the recombining electrons is completely transferred to the recombination centers: $P = p = 0.70$.

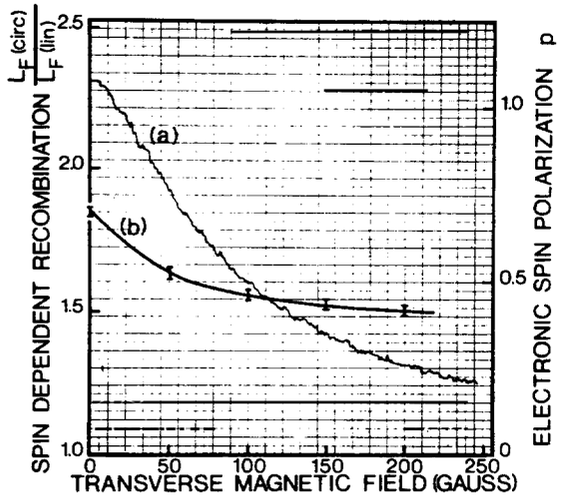

FIG. 2. Transverse magnetic field effect on the spin dependent recombination $L_F(\text{circ})/L_F(\text{lin})$ (a) and on the spin polarization p of the recombining electrons (b).

The well-known depolarizing effect of a transverse magnetic field[5] is shown in Fig. 2. It is seen that both the SDR photoluminescence ratio $L_F(\text{circ})/L_F(\text{lin})$ and the polarization of the recombining electrons decrease significantly in a field of 50 G. At higher fields, the electronic polarization tends to a limit of 0.40 whereas SDR disappears almost completely. One can then conclude that such a field is large enough to depolarize completely the recombination centers but not the recombining electrons, which means that the characteristic evolution time of the polarization is shorter for the electrons than for the centers. The remaining spin polarization of the electrons is simply the effect of polarized luminescence under circularly polarized excitation when no SDR is present. It disappears in a transverse field of a few kG. From equation (3) with $P = 0$, one finds $T_1/(T_1 + \tau_0) = 0.8$ as mentioned above. This transverse magnetic field effect shows here clearly the presence of two types of spins with different polarization evolution times.

It should be noted that the spin polarization of the centers is strongly dependent on the intensity of the excitation light. In fact, at low intensity, when the density of excited carriers is not large enough to polarize the recombining centers, we have verified that the intensity of the luminescence does not depend any more on the polarization of the luminescence light.

Finally, let us add that the same material has been studied by Ekimov and Safarov under comparable experimental conditions.[9] In particular, similar results on the depolarizing effect of a transverse magnetic field on the electronic polarization are reported, but no results on the total intensity are given. These authors interpret their data by a dependence of the electron spin relaxation time on the polarization of the nuclear spins of the lattice. This effect would not affect the total intensity of the luminescence and therefore does not explain our results. The fact that NMR of the lattice nuclei is detected by a variation of the polarization of the luminescence light[10] cannot be regarded as a proof of their model since a preliminary experiment at 1.7°K has shown that NMR can also be detected on the total intensity of the luminescence light.[11] This last result indicates that SDR is sensitive to the nuclear polarization and it seems therefore that the recombining centers are in strong hyperfine interaction with the nuclei of the lattice. Further experiments are in progress to elucidate this point and to obtain a clear understanding of the recombination process.

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On observe une recombinaison dépendant du spin dans $Ga_{0.6}Al_{0.4}As$ à 77°K sur l'intensité de la raie de photoluminescence des paires donneur–accepteur. Le temps de vie augmente de 2.3 quand les électrons photo-excités et les centres de recombinaison ont leurs spins orientés par pompage optique en lumière polarisée circulairement. L'orientation optique et la recombinaison dépendant du spin conduisent à une polarisation électronique de spin égale à 70% en régime permanent.