

Spin-dependent recombination in GaAs

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(Received 11 September 1979)

Spin-dependent recombination (SDR) has been studied via photoluminescence of samples grown by molecular-beam epitaxy (MBE) consisting of many thin GaAs wells separated by thin AlGaAs barriers. The photoluminescent-intensity enhancement with circularly polarized optical pumping is observed to be as much as 46% above the intensity level seen with linearly polarized pumping. Electron-spin polarizations as large as 0.84 have also been generated. Similar but smaller effects are seen in the emission from the relatively thick GaAs buffer layer which is grown by MBE prior to the growth of the superlattice. The SDR and electron polarization ρ of the multilayers and the GaAs buffer layer have been studied as a function of the pump intensity and a transverse magnetic field B (Voigt-geometry-Hanle effect). Both pulsed and cw laser pump sources were used. It is observed that SDR is absent at very low pump intensities and generally goes through a maximum as the pump intensity is increased. In all cases, the decrease of SDR with B is faster than that of ρ . It appears that these data cannot be adequately described quantitatively by the theory used earlier by others in a study of SDR in the donor-acceptor pair photoluminescence of $\text{Al}_{0.4}\text{Ga}_{0.6}\text{As}$.

INTRODUCTION

Many papers have appeared on optically induced luminescence and oriented electron spins in GaAs. However, to the authors' knowledge, this is the first report of photoluminescent studies of spin-dependent recombination in this material. Spin-dependent recombination (SDR) has been regarded¹ as a manifestation of the Pauli exclusion principle which prohibits two electrons with the same spin occupying the same orbital state. As in earlier work of $\text{Al}_{0.4}\text{Ga}_{0.6}\text{As}$,² it is assumed that there is present in the material a nonradiative recombination center which has an unpaired electron, i.e., it acts as a paramagnetic center. These centers, which are of unknown origin in this and earlier work, can become polarized parallel to the optically oriented majority-electron spins generated with circularly polarized light when the latter are present in sufficient number density. As a consequence, the minority-electron spins are opposite to that of the polarized unpaired electrons and can recombine nonradiatively via these centers. At the same time the effective lifetime of the majority-spin electrons is increased since they cannot recombine through these polarized centers. Therefore, this effect can lead to enhanced luminescence when pumping preferentially into one electron-spin state with circularly polarized light compared to that observed with linearly polarized light where both electron spins are generated equally.

These effects have been studied in this work with superlattice samples grown by molecular-beam epitaxy (MBE) consisting of many thin GaAs wells separated by thin AlGaAs barriers.³ Such multi-

layer structures can exhibit very large electron-spin polarizations at high densities and hence are well suited for studies of SDR.⁴

EXPERIMENTAL

The main emphasis here is on samples from a wafer which was designed to be an efficient source of optically generated polarized electrons.^{4,5} The structure consisted of a GaAs substrate ($[\text{Zn}] \sim 1.6 \times 10^{18} \text{ cm}^{-3}$) with a (100) growth surface, molecular-beam-epitaxy grown 1- μm -thick buffer layer of GaAs ($[\text{Be}] \sim 2 \times 10^{16} \text{ cm}^{-3}$), 106 periods of 63- \AA -wide GaAs wells and 52- \AA $\text{Al}_{0.21}\text{Ga}_{0.79}\text{As}$ barriers ($[\text{Be}] \sim 2 \times 10^{16} \text{ cm}^{-3}$), and finally, a 1000- \AA GaAs cap ($[\text{Be}] \sim 10^{19} \text{ cm}^{-3}$). The AlGaAs barriers are sufficiently narrow so that the structure is really a superlattice, i.e., there is coupling between the wells.^{3,6} Data obtained with a sample from which the thin GaAs cap had been removed lead to the conclusion that the GaAs cap has no significant influence on the results to be discussed.

For the optical pumping, two different sources were utilized, a pulsed tunable optical parametric oscillator (OPO) and a cw Kr^+ laser. In the case of the OPO, the linearly polarized incident light pulses, $\sim 0.3 \mu\text{sec}$ in duration, were sent through a 60-Hz electro-optic modulator driven by a sine wave whose amplitude was adjusted for a maximum retardation of the linear input polarization of $\pm \frac{1}{4}\lambda$ thus generating right and left circularly polarized light. With OPO pulse operation at $\sim 40 \text{ Hz}$, each successive input pulse was shifted by $\sim \frac{3}{2}$ cycles of the 60-Hz modulation. The generated photoluminescence was detected in the backward direction, $\sim 24^\circ$ off normal incidence, and

passed through a fixed circular polarization analyzer ($\frac{1}{4}\lambda$ wave plate plus linear polarizer) before entering a $\frac{1}{2}$ -m monochromator. Gated detection of the photoluminescence at the OPO pulse rate therefore gives the average photoluminescent signal I_{PL} generated for right (+) and left (-) handed inputs, linear (L) inputs, or whatever, depending upon the exact phase of the OPO pulses with respect to the modulator voltage. This signal was ratioed with that from a monitor of the pumping light, placed downstream from the modulator, to yield the magnitude of the SDR which is here defined as $R_{SD} = I_{PL}(\pm)/I_{PL}(L)$.

The photoluminescent signal at exactly one half the OPO repetition frequency (~ 20 Hz) was also detected and ratioed with the photoluminescent signal at the OPO repetition frequency. When the ~ 20 -Hz photoluminescence pulses are in phase with the positive or negative voltage peaks of the modulator voltage, this ratioed signal is $I_{PL}(\pm)/\frac{1}{2}[I_{PL}(+) + I_{PL}(-)]$. The deviation of this signal from unity is

$$\frac{2I_{PL}(\pm)}{I_{PL}(+) + I_{PL}(-)} - 1 = \frac{I_{PL}(+) - I_{PL}(-)}{I_{PL}(+) + I_{PL}(-)} \equiv \rho, \quad (1)$$

which is the polarization of the luminescence. Therefore, in this mode of operation, one can display R_{SD} and ρ simultaneously with a two-pen chart recorder.

For the cw source, the input polarization was chopped (+) and (-) mechanically at 480 Hz. The photoluminescent signal detected at 240 Hz was ratioed with that detected at 480 Hz to determine the polarization ρ . To obtain R_{SD} with the Kr^+ source, dc measurements were made using a calcite element to determine the polarization of the input beam.

The samples were mounted in a variable temperature Dewar, but most of the measurements were taken at $\leq 10^\circ K$. Both as-grown samples and samples annealed for 2 h at $800^\circ C$ were measured. It has been noted earlier that annealing the multilayer samples usually improves the photoluminescent efficiency.⁷ In this case annealing increased the photoluminescence from the GaAs buffer layer by ~ 2.5 and that from the multilayer by 4 to 9, the exact value depending upon the pump intensity. Annealing also usually results in a Stokes shift of the photoluminescence-intensity peak of the multilayer by a few meV.

RESULTS

Under the appropriate optical pumping conditions, photoluminescence from the wafer can be seen from the GaAs cap, the multilayer structure, the GaAs buffer, and from the GaAs substrate.

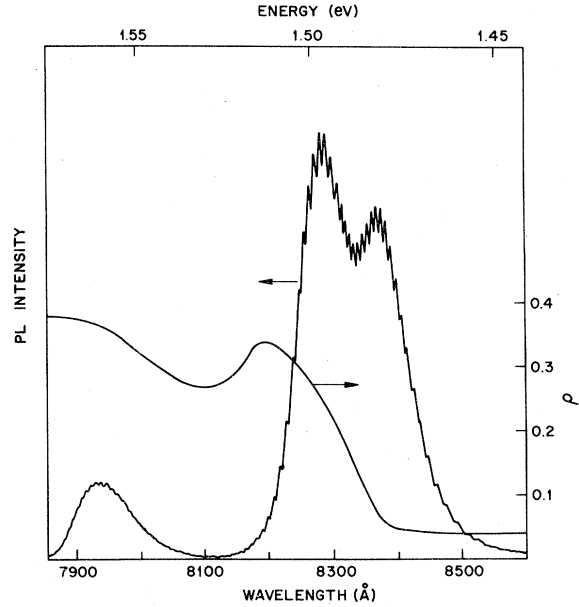


FIG. 1. Photoluminescent (PL) intensity and polarization versus wavelength for an annealed sample. The three intensity peaks are identified with the multilayers (7935 Å), GaAs buffer layer (8280 Å), and the GaAs substrate (8365 Å). The sample at $\sim 10^\circ K$ was pumped at 7632 Å with 12 kW/cm². The fine structure on the luminescence is due to spin-dependent recombination (SDR).

Figure 1 shows the photoluminescence intensity versus wavelength for the annealed sample when pumped by the OPO at 7632 Å with 12 kW/cm². Excitation spectra show that the three resolved peaks can be assigned as follows: GaAs wells (7935 Å), GaAs buffer (8280 Å), and the GaAs substrate (8365 Å). The fine structure on the plot represents SDR. However, in this plot its magnitude was somewhat time constant limited due to the fast scan speed used. In all cases $I_{PL}(\pm) > I_{PL}(L)$. For accurate measurements of R_{SD} or ρ neither the detected wavelength nor the pump was scanned.

Also shown in Fig. 1 is the polarization ρ vs λ obtained simultaneously with the photoluminescent spectrum. Note that except in the region of the substrate photoluminescent peak, ρ is greater than the maximum obtainable from bulk GaAs in the absence of SDR, namely, $\rho = 0.25$.⁸

The excitation spectrum for the multilayer photoluminescence and its ρ are shown in Fig. 2 for an as-grown sample. The monochromator was set on the peak of the multilayer luminescence 7910 Å (shifted 25 Å from the annealed-sample photoluminescence peak), and the input intensity 12 kW/cm² scanned in energy. Again, as shown in Fig. 2(b), SDR is clearly discernible with

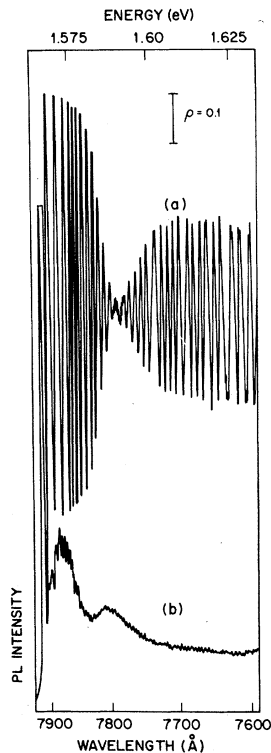


FIG. 2. The excitation spectrum for the multilayer polarization (a) and its photoluminescence intensity peak (b) for an as-grown sample. The $n=1$ electron to $n=1$ light and heavy-hole exciton transitions are clearly resolved at 7810 and 7885 Å, respectively. The magnitude of the polarization is given by the size of the vertical swings in the upper part of the figure. The fine structure in the photoluminescence near the heavy-hole transition is due to SDR.

$I_{PL}(\pm) > I_{PL}(L)$ and reaches a maximum at the intensity peak (7885 Å) which is the $n=1$ electron to $n=1$ heavy-hole exciton transition E_{1h} .^{3,4} Thus there is a 25-Å Stokes shift in the emission at the photoluminescence peak. The second peak (7810 Å) is the $n=1$ electron to $n=1$ light-hole exciton transition E_{1l} . The polarization ρ , given approximately by the magnitude of the vertical swings shown in the figure, is about 0.35 at the highest energy, decreases to ~ 0.03 at E_{1l} , and then increases to 0.84 at E_{1h} , the highest value reported for band-edge photoluminescence to date. These higher values may again be slightly time constant limited. However, the most important point here is that the photoluminescence polarization ρ becomes very large which therefore also indicates an equally large electron spin polarization.⁴

Figure 3 shows an excitation spectrum for the GaAs buffer layer of the annealed sample with the detector set at 8290 Å. The average pump inten-

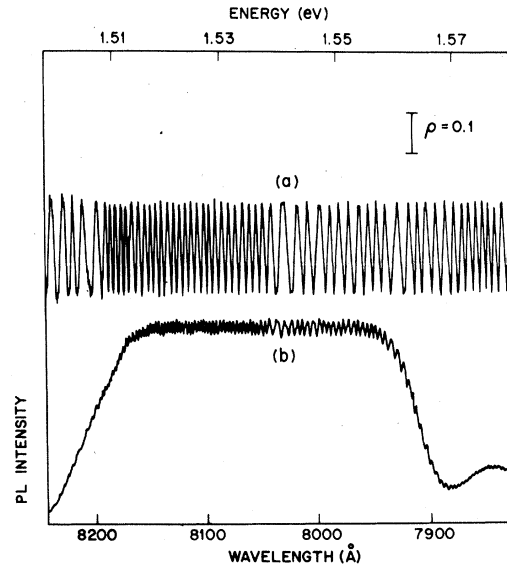


FIG. 3. The excitation spectrum for the GaAs buffer layer polarization (a) and its photoluminescence (b). The dip in the photoluminescence at 7885 Å is due to absorption by the heavy-hole exciton transition in the multilayers. While $\rho \sim 0.23$, the SDR is small and both are nearly constant. This sample had been annealed.

sity incident on the sample was about 5 kW/cm². However, the incident intensity decreased by a factor of about 3 from right to left, and that transmitted to the buffer layer increased by a factor of about 5 when the multilayer became transparent. The high-energy side of the figure reflects the absorption due to the multilayers and clearly shows the E_{1h} transition at 7885 Å. These data show conclusively that the photoluminescence at 8290 Å was generated deeper in the structure than the multilayer. The drop off in photoluminescence at ~ 8180 Å is due to the decrease in absorption that occurs when the pump wavelength passes through the GaAs buffer band edge. Note that ρ is ~ 0.23 and does not change as the excitation wavelength moved through the multilayer exciton transition. This shows that the large electron-spin polarization density at E_{1h} , ~ 0.8 as shown in Fig. 2, does not migrate into the wide GaAs buffer layer sufficiently to affect the net-spin polarization in the layer. The SDR is relatively constant at $R_{SD} \sim 1.07$. However, both ρ and R_{SD} vary with the photoluminescence wavelength detected and the pump intensity.

The excitation spectrum for the photoluminescence from the GaAs substrate (not shown) obtained with the detector set at 8375 Å shows R_{SD} a few percent above unity and a polarization ≤ 0.1 . These results are consistent with similar data obtained with the bare substrate material.

The polarization and SDR have been measured as a function of incident pump intensity I_p for both the multilayer and the GaAs buffer layer. The E_{1h} exciton transition was pumped for the multilayer while the buffer layer was excited at 8036 Å so that there would be no pump absorption in the multilayer. The monochromator was set at about 8260 Å for the buffer layer which is considered to represent best its photoluminescent spectrum over the I_p range investigated. For the multilayer, the detector was set at the wavelength of the peak of the photoluminescence which is not very sensitive to I_p .

The data for the annealed sample are shown in Fig. 4 for I_p from 0.2 to 200 kW/cm². Additional data obtained for both the annealed and unannealed samples with the Kr⁺ laser at 7525 Å show that $R_{SD} \sim 1.0$ for sufficiently small I_p . Both the ρ and R_{SD} for the multilayer increase somewhat with I_p and then fall off at the highest pump intensities. The highest value of R_{SD} is 1.46. The decreases of ρ and R_{SD} with I_p could be due to a saturation of the SDR centers. Also, band filling, which will be shown to occur at the highest I_p 's, could lead to a decrease of ρ . For the GaAs buffer layer, ρ and R_{SD} are roughly correlated and presumably drop off for sufficiently high I_p due to saturation effects. Note that for the GaAs buffer, ρ can be significantly larger than theory,⁸ 0.25, in some regions, an effect that is probably due to SDR.

The equivalent data for the as-grown sample are shown in Fig. 5. Since the photoluminescence due to the multilayer from this sample is less than that for the annealed sample by a factor of 4 to 9 (the factor depends on I_p) saturation and/or band filling effects should require higher values of I_p than for the annealed samples. This is the case as can be seen from Fig. 5. In fact, the SDR from

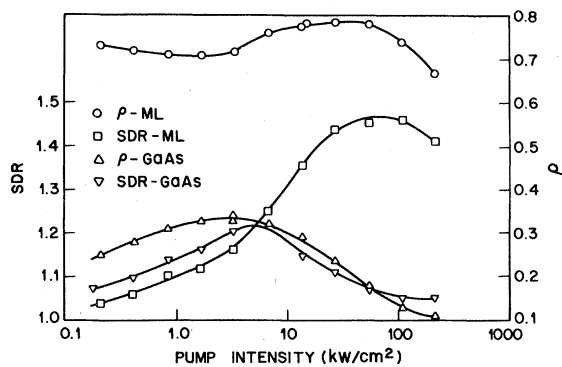


FIG. 4. The polarization ρ and spin-dependent recombination R_{SD} for the multilayer (ML) heavy-hole exciton photoluminescence peak and the GaAs buffer-layer photoluminescence versus pump intensity for an annealed sample.

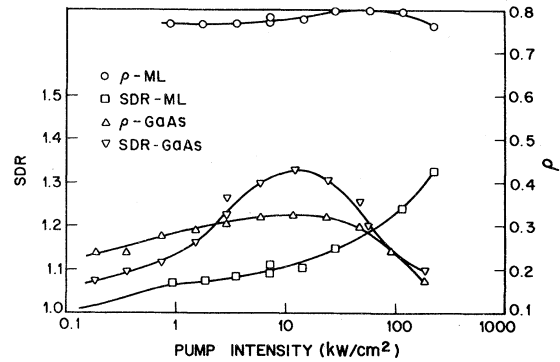


FIG. 5. The same as Fig. 4 but for an as-grown sample.

the multilayer does not appear to saturate at all over the I_p range covered. The photoluminescence from the buffer layer increased by about 2.5 upon annealing so that saturation of ρ and R_{SD} are observed with the as-grown sample but at higher intensities than with the annealed sample.

Band-filling effects in the GaAs wells are observed at the highest pump intensities with both the annealed and as-grown samples. As expected, the effects are largest with the annealed samples. The excitation spectrum for the buffer layer shown in Fig. 6 demonstrates the effect clearly. The monochromator was set at 8250 Å and the average incident pump intensity was ~ 200 kW/cm². At this intensity, E_{1h} is broadened but still resolved in the excitation spectrum of the multilayer photoluminescence (not shown). At the same time, the E_{1l} exciton transition is barely resolved in the multilayer photoluminescence but is clearly discernible in ρ due to the consequent reduction that occurs in ρ at this transition as shown for example in Fig. 2. However, neither E_{1h} nor E_{1l} is resolved in the buffer-layer excitation spectrum shown in Fig. 6. Throughout this plot, the local peaks in the photoluminescence are due to circularly polarized input polarizations. Band filling is pronounced in the photoluminescence where the multilayer becomes transparent. In this region, the GaAs buffer photoluminescence peaks represent increased transmission of the circular polarized pump through the multilayer. This increase results from the considerably higher electron Fermi level obtained with the highly polarized ($\rho \approx 0.7$) electrons generated with circularly polarized pumping over that obtained with linearly polarized pumping. These band-filling effects are only observed when pumping near the multilayer bandedge. The decrease in ρ shown in Fig. 6 as the multilayer becomes transparent is due to the resulting increased I_p on the GaAs buffer layer and

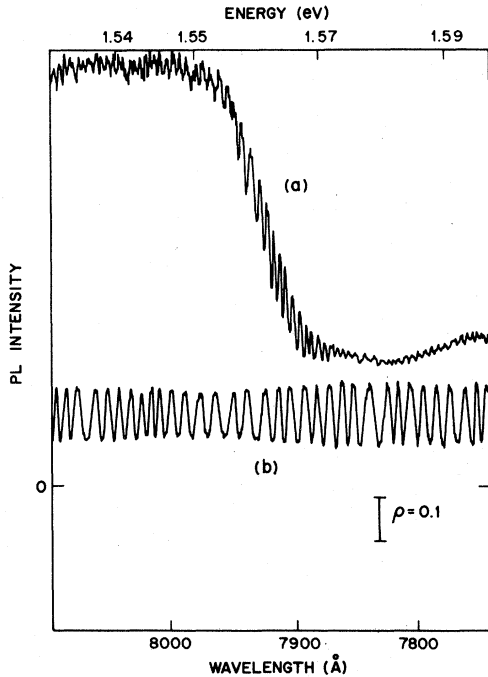


FIG. 6. The polarization of the photoluminescence of the GaAs buffer layer (a) and its intensity (b) versus photon excitation energy for an annealed sample pumped at 200 kW/cm^2 . The wide swings in the photoluminescence near 7910 \AA are due to band filling in the GaAs wells which is most pronounced with circularly polarized light.

is consistent with the data in Fig. 4. These band-filling effects must tend to reduce both ρ and R_{SD} in the photoluminescence near the E_{1h} transition of the multilayer.

The depolarization effects due to a magnetic field B transverse to the pump light propagation direction (Hanle effect)⁸ are shown in Fig. 7 for

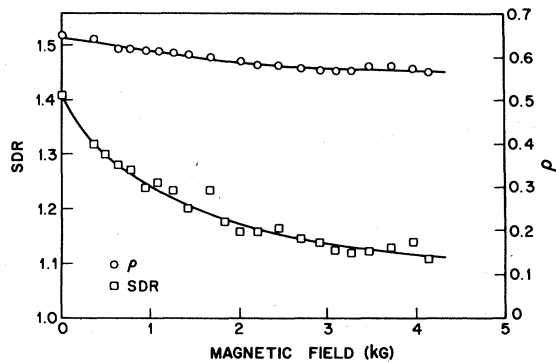


FIG. 7. Variations in the multilayer photoluminescence polarization ρ and spin-dependent recombination R_{SD} due to a transverse magnetic field for an annealed sample. The sample was pumped at the $n = 1$ heavy-hole exciton transition with 160 kW/cm^2 .

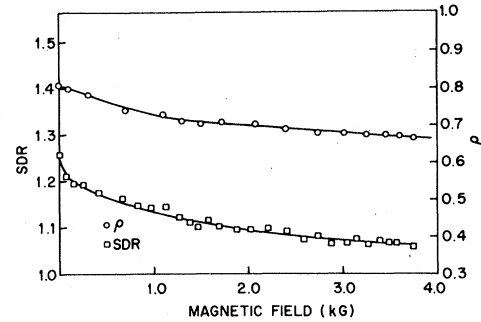


FIG. 8. Transverse-magnetic-field dependence of ρ and R_{SD} for an as-grown sample pumped at the $n = 1$ heavy-hole exciton with 98 kW/cm^2 .

the luminescence from the multilayer of the annealed sample with a pump intensity of 160 kW/cm^2 at E_{1h} . The polarization ρ decreases very little with field, suggesting that the electron lifetime and spin-relaxation time are both very short. In contrast, the SDR drops rapidly with field. Figure 8 shows similar data for an as-grown sample pumped at E_{1h} with 98 kW/cm^2 . At lower pump intensities, e.g., $\sim 10 \text{ kW/cm}^2$, the same trends are observed for the multilayer luminescence. Equivalent data for the GaAs buffer emission (8270 \AA) of an as-grown sample pumped with 13 kW/cm^2 at 8082 \AA are shown in Fig. 9. For the GaAs buffer layers, the R_{SD} again drops off faster than ρ , but the decrease of ρ with B can be as large as a factor of 3 or 4. When R_{SD} for the buffer layer drops below about 1.1, ρ is always less than theory, i.e., < 0.25 .⁸

A limited amount of SDR data have also been obtained with a second multilayer wafer. This wafer had 46 periods of 54-\AA GaAs wells and 52-\AA $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ barriers. Both sides of the multilayer were clad with $\sim 1 \mu\text{m}$ of $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ and all the MBE-grown layers were undoped. Some polarization data for this sample have been reported earlier.⁴ Electron-spin polarizations as large as

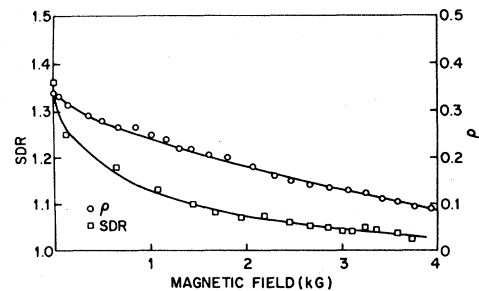


FIG. 9. Dependence of ρ and R_{SD} at 8270 \AA on a transverse magnetic field for the GaAs buffer layer of an as-grown sample. The sample was pumped at 8082 \AA with 13 kW/cm^2 .

0.7 were achieved, but the largest R_{SD} observed was 1.1 at ~ 80 kW/cm². Again the R_{SD} decreased with B more rapidly than ρ .

At low pump intensities with the Kr⁺ laser when no SDR is present, $I_p \lesssim 1$ W/cm², the Hanle plots of ρ^{-1} vs B^2 are very flat for the multilayer luminescence of the as-grown sample suggesting that no significant electron-spin relaxation occurs over the 0 to 4-kG magnetic field range explored. However, for all the other samples, plots of ρ^{-1} vs B^2 are observed to deviate from linearity at small B , i.e., $B \lesssim 1$ kG. These data are therefore "anomalous" in the sense that ρ^{-1} vs B^2 is not linear as required by the simple theory that says ρ should be Lorentzian in B .⁸ These samples are said to exhibit "Hanle anomalies."

DISCUSSION

The magnitude of the SDR observed with the principal wafer described herein is much larger than that seen by the authors in any other material. Examination of tens of samples grown both by MBE and liquid phase epitaxy shows that the usual level is a few percent or less. Subsequent wafers grown in approximately the same manner as the principal wafer failed to exhibit multilayer luminescence with either a very large ρ or a very large R_{SD} . However, the principal wafer was one of the first grown after recharging the MBE ovens which suggests that these effects may be influenced by an impurity whose concentration decreases with growth time. It is also observed⁹ that the photoluminescence intensity, i.e., efficiency, increases with time after the MBE ovens are recharged. What the two wafers discussed herein have in common for the multilayer part of the structure is a large spin polarization and at least some coupling between the wells. However, bulk GaAs samples heavily doped, $[Zn] \sim 10^{19}$ cm⁻³, can also exhibit large polarizations, i.e., close to the theoretical limit, but R_{SD} is generally at most only a few percent from unity.⁹

The SDR phenomena reported herein are reproducible in the sense that they are observed in all samples cleaved from a given wafer. Also, in the same sense, the effects of annealing are reproducible. In addition, it was verified that these SDR phenomena are not due to strains in the plane of the layers. Such strains can cause effects which look like SDR.⁹ The present effects are found to be isotopic about the normal to the plane of the layers.

Weisbuch and Lampel² (WL) derive expressions for both ρ and R_{SD} in terms of the polarization $P \equiv (N_+ - N_-)/(N_+ + N_-)$ of the paramagnetic centers N and the electron polarization

$$\rho_1 = p_i / (1 + \tau / \tau_s), \quad (2)$$

where τ and τ_s are the electron lifetime and spin-relaxation time, respectively, in the absence of SDR. The quantity p_i is determined by the band structure and equals 0.5 for bulk GaAs and 1.0 for the E_{1h} transition when pumped in multilayer structures. The values of ρ_1 and hence $1 + \tau / \tau_s$ were estimated by WL from the magnetic field dependence of ρ and R_{SD} . Then the center polarization P can be estimated from the WL relations:

$$R_{SD} = [1 + (1 + \tau / \tau_s) p_i P] [1 - (1 + \tau / \tau_s) P^2]^{-1} \quad (3)$$

and

$$2\rho = (1 + \tau / \tau_s) (p_i + P) [(1 + \tau / \tau_s) p_i P]^{-1}, \quad (4)$$

which apply at $B=0$. The WL data on donor-acceptor pair photoluminescence of Al_{0.4}Ga_{0.6}As give $R_{SD} = 2.3$, $\rho_1 = 0.8$, and $2\rho = 0.7$ which are consistent with $P \sim 0.6-0.7$ in Eqs. (3) and (4).

While similar analyses of the present data have been obtained and will be discussed, the results must be viewed with caution. The WL approach requires that the nonlinear behavior of ρ^{-1} vs B^2 for small B results from SDR. However, of all the samples studied in this investigation only the multilayer luminescence of the as-grown samples from the principal wafer failed to exhibit a Hanle anomaly when there was no SDR at low pump intensities. Another difficulty is that the WL model assumes the photoluminescence intensity is a linear function of the pump intensity I_p . In the present case the photoluminescence peak from the multilayer is observed to vary as $I_p^{1.4}$ up to about 10 kW/cm² after which it tends to saturate, i.e., it actually goes sublinear in I_p . The I_p dependence of the photoluminescence from the GaAs buffer layer is not so easy to ascertain due to the overlapping substrate photoluminescence. However, the photoluminescence intensities measured at a fixed wavelength near the GaAs buffer photoluminescence peak again increases superlinearly with I_p over much of the region of interest here. In the earlier work by WL, the pump dependence of the photoluminescence intensity was not given.

In any event, the WL-type analysis has been tried with the present data. For these analyses, plots of ρ^{-1} vs B^2 are extrapolated to $B=0$ to yield ρ_1 which turns out to be unity for the GaAs buffer material when the SDR is large. Thus, although $P \sim 0.3$ and 0.2 would seem to fit the $B=0$ buffer-layer data for the as-grown and annealed samples, respectively, the values of P are suspect for the reasons given above. Again the multilayer data in Fig. 7 yield $P \sim 0.4$, but this sample always shows a Hanle anomaly for the multilayer polarization.

Several Hanle runs have been made on the multilayer photoluminescence of an as-grown sample from the principal wafer with sufficiently high pumping intensities to generate an R_{SD} of ~ 1.2 . One of these runs is shown in Fig. 8. Recall that this sample does not exhibit a Hanle anomaly at very low I_p , where $R_{SD} = 1.0$. The various data on R_{SD} yield $P \sim 0.3$ from Eq. (3), while the ρ data give $P \sim 1$ from Eq. (4). Thus it does not appear that any of the present data can be described satisfactorily by the WL approach.

CONCLUSIONS

This extensive study of SDR in multilayers consisting of many thin GaAs wells separated by thin $\text{Al}_x\text{Ga}_{1-x}\text{As}$ barriers, and in essentially bulk GaAs, demonstrates that this effect can be far from trivial in MBE-grown material. The data show that the biggest effects are observed with layers that exhibit a large electron-spin polarization, which we find, in the case of multilayers, correlates with low levels of photoluminescence, i.e., a low quantum efficiency. The pump intensity dependence of the SDR is reasonable in the sense that $R_{SD} \rightarrow 1.0$ as $I_p \rightarrow 0$ and saturation effects can be ob-

served with sufficiently high pump intensities. The decrease of ρ and R_{SD} with a transverse magnetic field is qualitatively as expected, e.g., ρ is always less than or equal to the maximum theoretical value for $R_{SD} \rightarrow 1.0$. However, none of the data on the various samples can be described quantitatively in terms of the existing theory, namely that of Weisbuch and Lampel.² Complications with the present multilayer data involve the observed superlinear dependence of the photoluminescence on I_p and in all but one case, the low field Hanle anomalies. An added complication with the GaAs buffer-layer data may arise from the fact that the buffer-layer photoluminescence is not completely resolved from that of the GaAs substrate.

ACKNOWLEDGMENTS

The authors would like to thank Dr. D. A. Kleinman for many useful and stimulating discussions on the subject of this paper, and Dr. A. C. Gossard for the design and assistance in the growth of the two MBE-grown wafers used in this study. Several informative discussions with Dr. C. Weisbuch are also gratefully acknowledged.

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