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Mechanisms for the optically detected magnetic resonance background signal in epitaxial GaAs

F. P. Wang, B. Monemar, and M. Ahlström
Department of Physics and Measurement Technology, Linköping University, S-581 83 Linköping, Sweden

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The mechanisms for the usually very strong background signal in optically detected magnetic resonance (ODMR) experiments have been studied in high-purity epitaxial GaAs layers. The optically excited carriers are heated by the microwave field, via both cyclotron resonance and other microwave absorption processes involving free-carrier transitions within a Landau subband. The main mechanism for the change of photoluminescence (PL) by the hot carriers in the temperature range 4–10 K studied in this work is concluded to be impact ionization of shallow bound excitons (BE's) and shallow donors by hot free carriers, resulting in a quenching of the shallow PL emissions related to BE's and donor-acceptor pairs. These processes explain the microwave-induced change of PL intensity (the ODMR background signal).

Optical detection of magnetic resonance (ODMR) is a powerful technique for studying the identity and electronic structure of defects in semiconductors.[1] Strong and very broad nonresonant background signals are often observed in ODMR experiments, however, which has limited the application of ODMR, especially for GaAs.[2] The mechanism for such background signals was first discussed by Romestain and Weisbuch[3] and is still a controversy.[4-6] Possible mechanisms are impact ionization of bound excitons (BE's) by hot carriers, nonradiative decay processes which compete for the concentration of free carriers, bolometric effects,[7] and various temperature dependences of, e.g., photoluminescence (PL) line shape[3] or capture rates.[8]

In this Rapid Communication we report a detailed study of the ODMR background signal in high-purity epitaxial GaAs. We conclude that impact ionization of shallow bound excitons (BE's) and shallow donors via hot carriers accelerated by the microwave field is the dominating physical mechanism for the background signal. Compared to the results of a recent similar study for silicon,[9] the ODMR background is important at much lower microwave-power levels (or electric fields) in GaAs. Further, donor breakdown is an unimportant mechanism in the silicon case,[9] while for GaAs a clear demonstration of donor breakdown in a microwave field is presented in this work.

Photoluminescence was excited with an Ar^+ 514-nm laser line and detected with a cooled North Coast EO-817 Ge detector. For ODMR background studies, a modified Bruker 200-SRC ESR spectrometer was used, equipped with a cylindrical cavity operating in the TE_{011} mode, with optical access from all directions. The sample (about $2\times2\times0.4$ mm^3) was always placed in the center of the cavity, and was cooled with a continuous He flow Oxford Instruments ESR10 cryostat, where the sample temperature could be regulated from 4 K and up. A lock-in amplifier was employed to measure the magnitude of the microwave-induced change of PL intensity. A Jobin-Yvon 0.25-m grating monochromator was used on the detection side to obtain the spectral dependence of the microwave-induced signals. The samples used in this study were from nominally undoped liquid-phase epitaxy (LPE) GaAs wafers, with a net doping (originating from C and Si contamination) in the low 10^{14}-cm^{-3} range in the rather thick (20 μm) epitaxial layers. Metalorganic chemical-vapor deposition (MOCVD) grown n-type layers with a doping of about $n=2\times10^{15}$ cm^{-3} (Si doped) were also investigated for comparison.

Low-temperature PL spectra of LPE GaAs samples have been reported previously[10] and agree with those observed in this work. The spectra show a neutral-donor BE (D^0X) line at 1.5141 eV, the neutral-donor to free-hole (D^0h) or exciton-bound to ionized-donor (D^+X) line at 1.5133 eV, the neutral-acceptor BE (A^0X) line at 1.5124 eV, and further donor-acceptor pair (DAP) and free to bound (FB) emissions with the following energies: Si DAP, 1.482 eV; Si FB, 1.485 eV; C DAP, 1.490 eV; and C FB, 1.495 eV.

Figure 1 shows ODMR background spectra measured as the total change of the near-band-gap PL intensity with magnetic field in the Faraday configuration for different microwave-power levels (chopped at 6 kHz). The background signal was very large, about an 80% change of the total PL intensity was typically observed in the high-microwave-power case. Spin-dependent effects can be ruled out since no spin-resonance signal could be observed in the spectrum. Any mechanism responsible for this ODMR background would then be expected to relate to free-carrier heating by the microwave radiation.

An interesting observation from Fig. 1 is that the ODMR background signal is not zero even at the highest magnetic fields used in the experiments, that is, at 1 T. Cyclotron resonance (CR) absorption of microwaves, causing heating of free carriers, is not effective in a magnetic field as high as 1 T at 9 GHz. This means that CR is apparently not the only mechanism by which free carriers are heated in these experiments. Other mechanisms responsible for heating the carriers could be scattering processes, causing transitions of carriers to higher-energy states, but within the same Landau subband,[11] e.g., with absorption or emission of a phonon to fulfill the momen-

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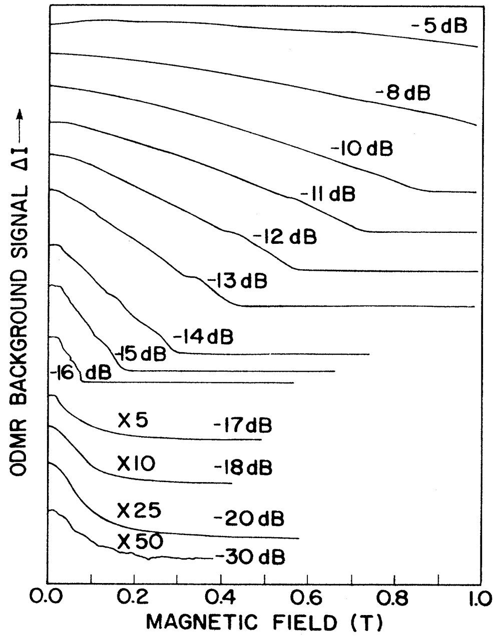

FIG. 1. The ODMR background spectrum for microwave-power levels ranging from -5 to -30 dB (0 dB corresponds to 577 mW), under stationary laser excitation (0.5 mW Ar^+ 514-nm laser line), measured as the change of the total PL intensity induced by the chopped microwave field (at 6 kHz) vs magnetic field in the Faraday configuration. The (LPE GaAs) sample surface normal was oriented about 40° away from the magnetic field direction.

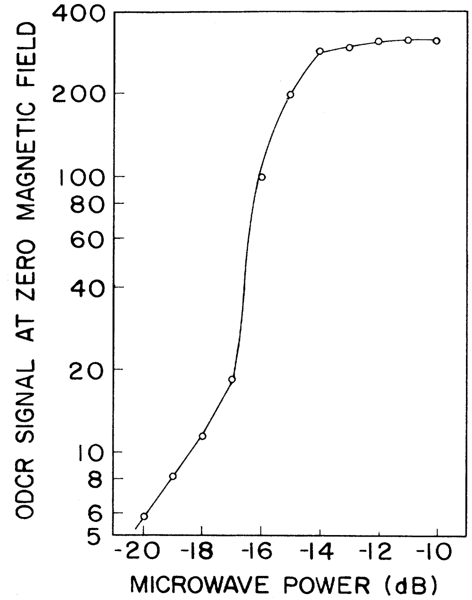

FIG. 2. The ODCR signal at zero magnetic field vs microwave-power level, taken from spectra such as those shown in Fig. 1. The laser excitation was stationary (0.5 mW), and the microwaves were chopped at 6 kHz. The amplitude of the microwave-induced change of the PL signal was measured. The (LPE GaAs) sample temperature was about 4 K without microwaves applied.

tum-conservation condition.[12] If this magnetic-field-independent contribution is removed, the remaining ODMR background signal is expected to be due to a broadened cyclotron resonance of free carriers (in the limit of $\omega\tau_c < 1$, where τ_c is the carrier scattering time) and can be denoted as the optically detected cyclotron resonance (ODCR) signal.

Figure 2 shows this ODCR signal versus microwave power. A threshold is clearly seen around the -17-dB level of relative microwave power, where the ODCR signal is rapidly increased more than 1 order of magnitude with increased microwave power. This threshold suggests that impact ionization is the mechanism responsible for the background signal.[13] The free-carrier heating by the microwave power leads to an increasing carrier density, due to impact ionization of shallow excitons and neutral donors. At very low microwave power, a linear function between the ODCR signal and the cyclotron resonance absorption would be expected, as is also observed below the -17-dB level of microwave power in Fig. 2. Above -17 dB, the impact processes of neutral donors dominate above 4.2 K, resulting in a rapid increase of the ODCR signal.

Impact ionization is expected at rather low electric fields for shallow bound excitons and neutral donors in GaAs, and has previously been observed by monitoring the PL intensity in a dc electric field in n-type GaAs.[13] These data have shown a complete quenching of free-exciton and bound-exciton PL at 2–4 V/cm. Comparing the data for donor breakdown in Fig. 2 with these previous data for PL quenching in GaAs in a dc electric field,[13] the -17-dB level of relative microwave power corresponds to an electric field of about 3 V/cm in the sample region.

The threshold microwave power (Fig. 2) was observed to be sample dependent, which could be explained as partly due to differences in carrier mobility, and partly due to a scatter in the impact-ionization cross section resulting from local sample inhomogeneities. Comparing the microwave threshold power in a range of samples, both n-type and p-type LPE samples show that the major factor determining the threshold power is the sample quality, i.e., doping level and carrier mobility. This result is only consistent with the impact-ionization mechanism. A low carrier mobility requires a higher field to reach the threshold kinetic energy for carriers to ionize BE's and neutral donors, leading to electrical breakdown. Consistent with the above observations, the MOCVD samples, which have a much higher doping level ($N_D - N_A \approx 2\times 10^{15}$ cm^{-3}) show a higher ODCR threshold, about 10 dB.

Since the electron mobility in GaAs is always very much larger than the hole mobility, the electrons will gain energy from the microwave field much faster than the holes. Therefore, the ODMR background signal is expected to be strongly dominated by electrons, and the effects discussed here are therefore regarded as exclusively related to free electrons.

In Fig. 3 we present the spectral dependence of the ODMR background signal at 4 K, for different microwave-power levels (chopped at 6 kHz). Only the in-phase component of the synchronous PL signal has been moni-

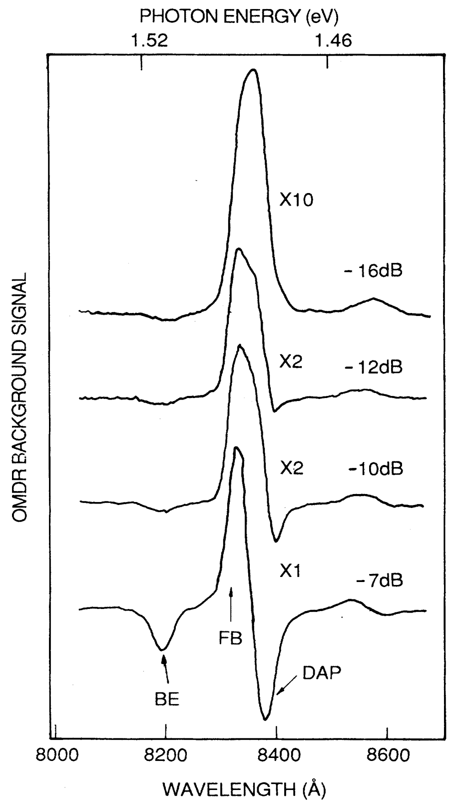

FIG. 3. The spectral dependence of the ODMR background signal for an LPE GaAs sample measured in the low-resolution ODMR setup at a lowest temperature of about 4 K (when no microwave field is applied). Relative microwave-power levels (chopped at 6 kHz) ranging from −7 to −16 dB were employed. No magnetic field was applied.

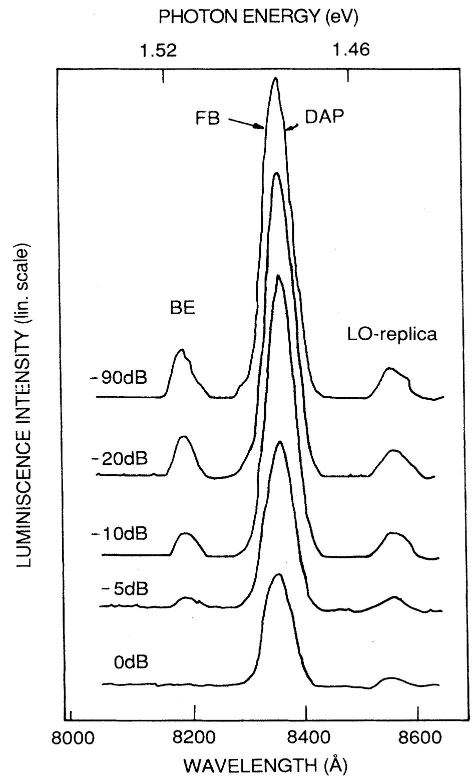

FIG. 4. Photoluminescence spectrum (measured in phase with chopped laser pulses) for a LPE GaAs sample under simultaneous microwave-power excitation for different microwave-power levels (chopped at 6 kHz). The laser excitation power was 4 mW.

tored, minimizing the possible lattice heating effect, as will be discussed below. It is clearly seen in Fig. 3 that both the shallow BE and the DAP emissions are quenched in the high-microwave-power case by the impact processes, while the FB emission is enhanced, due to the increased free-carrier density. A slightly different threshold for BE and DAP emissions, respectively, is evidenced in Fig. 3, as expected since the binding energy of the donors is slightly larger than that of the BE's. These results directly show that impact ionization of both BE's and neutral donors are responsible for the ODMR background signal.

In order to minimize the effects due to lattice heating, very low laser power of about 0.5 mW has been used for the excitation, and a lock-in amplifier is used to pick out only the synchronously changed part of the PL signal at a quite high microwave chopping frequency (6 kHz). In this way the bolometric effect (known to be a slow process) has been minimized, but nevertheless some thermal accumulation during the pulse can probably not be avoided in the ODMR experiment. This is directly demonstrated in Fig. 4, where the ordinary PL spectrum at a higher (4 mW) laser-power level under simultaneous microwave-power excitation for different microwave-power levels is presented. For higher microwave power, heating of the lattice in this case apparently leads to thermal quenching of the PL intensity. At the 0-dB level of microwave power (577 mW), the shallow BE emissions have been completely quenched. The lattice temperature dependence of the PL spectrum was measured separately in a high-resolution setup. Comparing the obtained temperature-dependent spectra in these measurements with the ones in Fig. 4, it was obvious that the lattice temperature of the sample could have reached about 10 K at the 0-dB microwave-power level (Fig. 4). These results indicate that even with the lower level of laser power (0.5 mW) the sample temperature is somewhat dependent on the microwave-power level. This does not mean that the lattice heating effect has any major influence on the measured ODMR background signal in this work, however, since only the synchronously changing part of the PL is monitored in the ODMR experiments. The time-resolved ODMR background signal has also been measured with a boxcar-averaging system, and the signal shows a fast response with the applied microwave pulse, which argues against any important contributions from bolometric effects.

In conclusion, we report an investigation of the frequently observed strong ODMR background signal in GaAs. We show that impact ionization of shallow bound excitons and shallow donors by hot electrons is the dominating mechanism responsible for this background signal, which is a severe disadvantage in investigating spin resonance of defects. Properly understood, on the other hand, it allows interesting studies of the luminescence processes in the presence of microwaves and magnetic fields, not readily obtainable otherwise.

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