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Full-spectrum optically detected resonance (ODR) spectroscopy of GaAs/AlGaAs quantum wells[1]

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Abstract

Resonant magneto-absorption of far-infrared (FIR) laser radiation by neutral and negatively charged donors and free electrons, and the mechanisms of coupling of the power absorbed to various photoluminescence recombination paths in a Si-doped GaAs/AlGaAs multiple-quantum-well structure was studied by optically detected resonance (ODR) spectroscopy. A sensitive charge-coupled-device detection scheme was used to record complete PL/ODR spectra at high resolution with good signal-to-noise. The rich and complex ODR spectra were analyzed by a line-fitting procedure. Results on the neutral donor 1s–$2p_+$ transition show that at low FIR laster intensities, the recombination is modified by processes that do not involve carrier heating. At high FIR laser intensities, carrier heating effects dominate.

Keywords: FIR spectroscopy; Photoluminescence; Impurities

Optically detected resonance (ODR) spectroscopy combines visible/near-infrared photoluminescence (PL) with far-infrared (FIR) methods [1–6]. In most implementations a chopped, monochromatic FIR laser beam excites electronic transitions whose energies are tuned into resonance by an applied magnetic field; changes in the PL, simultaneously excited by a visible laser with photon energy greater than the effective band gap of the structure under investigation, are synchronously detected. Optically detected resonance spectroscopy possesses several distinct advantages over conventional FIR techniques, including sensitivity, and spectral specificity of both the FIR absorption and the recombination. The latter offers the possibility of obtaining detailed information about energy transfer mechanisms from the various internal transitions excited by the FIR beam to the different recombination channels, as well as identification of unknown features in either the FIR or PL spectra. In order to realize this promise, a more complete understanding of the ODR mechanisms is needed. In most previous ODR studies small changes in intensity of PL over some fairly wide spectral window centered on one or more PL features were monitored as a function of applied magnetic field. The use of CCD-array detection allows sufficient sensitivity to obtain

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the entire ODR spectrum at high resolution without loss of spectral information [7,8]. Thus, detailed changes of particular PL features correlated with FIR resonant absorption can be monitored.

The sample used was an MBE-grown GaAs/$Al_{0.3}Ga_{0.7}As$ MQW structure (six repetitions of 210 Å wells and 150 Å barriers) doped over the central one-third of the wells with Si donors ($N_D = 1 \times 10^{16}\,cm^{-3}$). This sample has been studied extensively [4,9,10]. Similar samples grown under the same conditions show partial compensation (25–30%) of the donors [11]. In earlier ODR studies of this sample [4] we observed an apparent increase in the free heavy-hole exciton (X) PL and a decrease in the region of the donor-bound exciton (D^0–X) PL at fields corresponding to the D^0 1s–$2p_+$ transition, the singlet transition of the negative donor ion (D^-), and electron CR. The ODR signals were attributed primarily to resonant carrier heating and competing recombination processes; more energetic free electrons lead to increased ionization of the donor-bound exciton, and a concomitant increase in the free exciton (X) recombination. To elucidate the ODR mechanisms in more detail, high-resolution measurements at 2 K were carried out at zero magnetic field, and at fields corresponding to the D^0 1s–$2p_+$ transition, the D^--singlet transition, and electron cyclotron resonance, while varying FIR power at low visible laser excitation power.

The experimental setup incorporates CCD-array detection into the arrangement reported previously [4,7]. The FIR laser was operated in CW mode and chopped at 1 Hz by an external shutter under the control of the computer operating the CCD array. Photoluminescence was excited with the 632.8 nm line from a 0.5 mW HeNe laser or the 514.7 nm line of a 1 W argon ion laser. Complete PL spectra were recorded and stored for each cycle of the shutter as a function of magnetic field (while sweeping the field slowly), and at several fixed fields corresponding to the positions of FIR resonances. The HeNe and FIR laser power dependence, and the temperature dependence (between 1.6 and 16 K) of the PL were investigated in detail at these fixed magnetic fields. The ODR spectrum is the difference between the PL spectrum with the shutter open (FIR on PL) and the PL spectrum with the shutter closed (dark PL). Many spectra (up to 800) could be averaged to record weak signals with

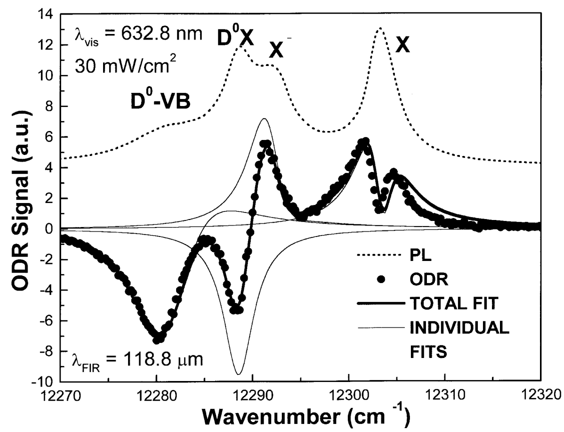

Fig. 1. Photoluminescence (upper panel) and ODR (lower panel) at 2 K and 2 T with the 118.8 μm line of the FIR laser at intermediate power. The various PL lines are labeled. Thin solid lines are fits to the individual contributions to the ODR signal as described in the text. The thick solid line is the total fit.

good signal-to-noise. The relative FIR laser power was measured in situ by a GaAs detector located below the sample at liquid-helium temperature. Over the range of powers available, the detector response was approximately linear. For simplicity, we concentrate in this report on the low-temperature data at low visible laser (HeNe) excitation intensities, for which the carrier heating effects due to the visible laser excitation are minimized, and the various physical mechanisms are easiest to distinguish. A more detailed and extensive description of the experiments will be presented elsewhere.

The upper trace of Fig. 1 shows the PL spectrum at $B = 2$ T, the resonant field for the 1s–$2p_+$ neutral donor transition. The four PL features are the free exciton (X), the negatively charged exciton (X^-), the neutral-donor-bound exciton (D^0–X), and the donor-to-valence-band transition (D^0–VB). From Fig. 1, it is clear that features in the ODR signal are sharper than in the direct PL, and the resulting spectrum is much richer and more complex. In spite of this complexity, these data can be fit very well by considering changes in not only the magnitude (or area) of a PL feature, but also (or exclusively) the position and/or the width of the feature induced by absorption of the FIR radiation. From these fits the underlying dependence of individual PL lines on, e.g., FIR laser power can be extracted.

The fitting procedure mimics the way in which the data are taken. First, the dark PL spectrum is fitted by the sum of four Lorentzian lines of varying area, position, and width. (This 4 line spectrum is adequate for low magnetic fields; however, at fields above 4 T, splittings and crossings of the excitonic features occur which make the spectrum even more complex.) The Lorentzian line shapes are expected to be reasonable for the three excitonic features. The line shape of the D^0–VB recombination is not known; it depends on the temperature and the distribution of neutral donors in the wells, as well as on possible hole localization due to lateral potential fluctuations. Thus, the Lorentzian fits to this line cannot be given physical meaning, and are rather used primarily to remove (in part) the influence of this recombination on the excitonic features.

The simulated ODR signal originating from each PL feature is obtained as the *change* in area, position and width from the dark PL Lorentzian line. This fit also determines the FIR-on PL which is the sum of the dark PL plus the (typically small) change (ODR signal). The resulting spectra are extremely sensitive to small changes in line shape and position, and the particular line shape provides clues as to the physical mechanism(s) leading to the changes in dark PL due to resonant absorption of the FIR light. An example of the fits is also given in Fig. 1 at an intermediate FIR power. The negative ODR signal in the region of the D^0–VB recombination results from a small shift and broadening of the fitted Lorentzian. The change in PL at the D^0–X line is primarily a *decrease* in integrated intensity; the X^- ODR signal corresponds to an *increase* in integrated intensity, a decrease in width and a slight shift to higher photon energies; and finally, the heavy-hole free-exciton PL (X) increases in integrated intensity, increases in width, and shifts slightly to higher photon energies, giving rise to the odd looking, double-peaked structure at this FIR power.

In order to simplify further discussion and to provide detail about the results of the fitting and the physical interpretation, we focus on the FIR laser power dependence of the ODR signal associated with the D^0 1s–2p$_+$ absorption feature at 118.8 μm ($B = 2$ T). The FIR power dependence is shown in Fig. 2. The ODR signal is normalized by the FIR power in each case. The most striking changes in the ODR signal with FIR power occur near the heavy-hole free exciton and the X^- PL lines. The X ODR signal changes

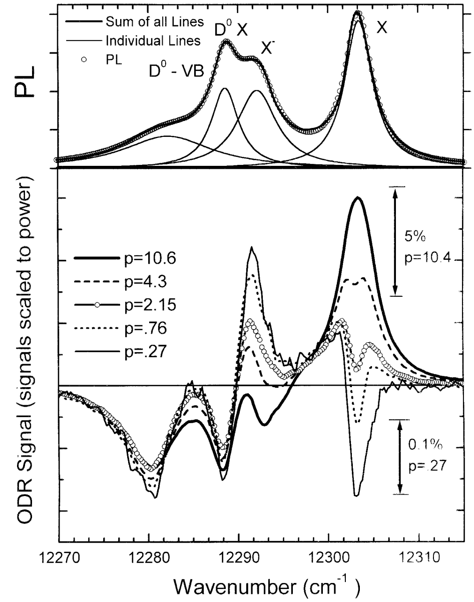

Fig. 2. Far-infrared power dependence of the ODR signals at 2 K and 2 T with the 118.8 μm line. The upper panel shows the PL. Thin solid lines shows fits to the individual features. The thick solid line is the total fit to the PL spectrum.

from a simple, entirely positive line at the highest FIR power through a series of double-peaked structures to a predominantly negative-going, but asymmetric feature at the lowest FIR power. The contribution from the X^- PL line maintains its shape, but changes from a positive-going, but overall negative feature, to an entirely positive feature as the intensity is decreased. The normalized change in the D^0–VB recombination is very weakly dependent on FIR power.

The results of fitting the ODR signals by the procedure described above are shown in Fig. 3 for the three excitonic recombination lines. The change in area (integrated intensity) of the Lorentzian lines used to fit the ODR signal associated with the three excitonic lines is plotted as a function of FIR power. A good fit to the ODR signal from the D^0–X line is obtained by *decreasing* the integrated intensity of the PL, and this reduction in integrated intensity *increases* with FIR power. There are also small changes in the line width and position of this feature that are not considered to be significant. On the other hand, in order to fit the X^- ODR signal, changes in three parameters are necessary; the integrated intensity increases, the width

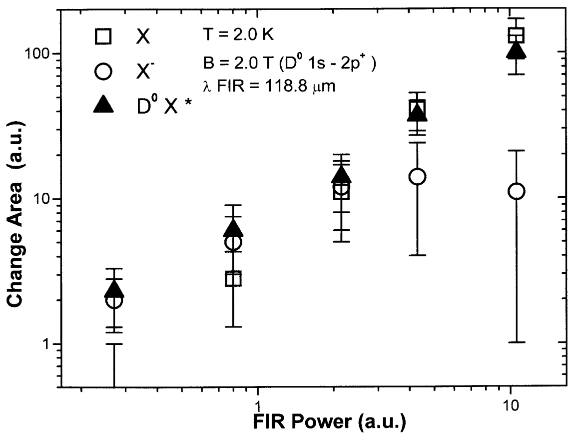

Fig. 3. Change in area (proportional to integrated intensity) of the various excitonic PL lines versus FIR power in arbitrary units. The (negative) *decrease* in area for the D^0–X feature is plotted as positive for ease of comparison with the other two contributions.

decreases, and the position increases slightly. The increase in integrated intensity saturates, and over a wide range is approximately independent of FIR power. At the highest power the dominant effect that contributes to the ODR signal in this frequency region is a shift (to higher frequency) of the PL line. Finally, the ODR signal associated with the free heavy-hole exciton (X) is fitted well by an increase in integrated intensity that is *strongly* dependent on FIR power (zero within the error at low power, and increasing faster than linearly). At the lowest power, the odd looking line shape in Fig. 2 results from a very small shift and broadening of the PL line with no change in integrated intensity.

The observed changes at low FIR power are attributed to photo-thermal ionization of neutral donors ($D^0 \rightarrow D^+ + e^-$) with no significant electron heating. Under the $30\,\mathrm{mW/cm^{-2}}$ visible laser excitation, a large fraction of the compensated donors (away from the well centers) are neutralized. Absorption of a FIR photon in the D^0 1s–$2p_+$ transition leaves the donor in the excited $2p_+$ state, and thermal ionization from this state to the continuum, or to the lowest Landau level at finite fields, is very efficient [12]. The electron in the conduction band Landau level is rather long lived [13,14]. Only Auger processes involving relaxation of electrons back to the donor ground state and excitation of other electrons to higher conduction band states can provide energy to the free-carrier system. Such processes are more effective at higher electron densities [15]. We have estimated the carrier heating by comparing the change in PL (the ODR signal) under resonant absorption conditions with the change in PL due to lattice heating, and find that under the highest FIR power conditions in the experiment the changes imply a carrier temperature rise of about 0.5 K.

At 2 T the photothermal process involving the 1s–$2p_+$ transition primarily ionizes donors at the well centers, with little or no carrier heating at low FIR power. The decrease in integrated intensity of the PL associated with the D^0–X line results from this photothermal ionization, which decreases the density of neutral donors available for recombination. The integrated intensity of the X^- PL increases since there are additional electrons available to create negatively charged excitons. The only at the lowest FIR power in the free heavy-hole exciton (X) line is a small broadening and a very small ($\cong 0.002\,\mathrm{cm^{-1}}$) shift to higher frequencies. This is attributed to a small modification of the lateral fluctuating potential seen by the free excitons due to the creation of a small density of D^+ ions by the photothermal process, and is indicative of the extremely high sensitivity of this technique.

At higher FIR power, the increasing density of free electrons leads to an increase in the Auger scattering rate, and a redistribution of the electrons among the Landau levels, i.e. electron heating. The continuing decrease in the D^0–X integrated intensity with FIR power is ascribed to a combination of further decrease in the density of neutral donors due to the photothermal process, and resonant carrier heating leading to ionization of the donor-bound excitons. Both effects lead to a decrease in the PL integrated intensity of the donor-bound exciton, and a concomitant increase in the integrated intensity of the competing free exciton (X) PL. The integrated intensity of the negatively charged exciton (X^-) PL increases initially, and saturates for FIR power >2 in Fig. 3 [16]. The initial increase is due to additional excess electrons from the photothermal ionization creating additional X^- excitons, while the saturation (and incipient decrease at the highest FIR power) appears to result from carrier heating which dissociates the weakly bound second electron. The ODR signal in Fig. 2 at the highest FIR power in the region of the X^- PL is almost entirely due to a *shift* of the peak to lower photon energy. Thus at the highest FIR power the intensity dominant effects in the ODR signal are competition between free heavy-hole exciton (X) recombination and the D^0–X recombination.

Similar complex line shapes and processes are involved with the ODR signals associated with resonant absorption at the D^--singlet and electron CR. At the highest powers, however, the ODR signals in all cases result predominantly from carrier heating by the resonant absorption. It is only at the lowest excitation intensities and the lowest FIR intensities that processes other than carrier heating are clearly observable and dominate the ODR signals.

In conclusion, we have shown that ODR spectroscopy with CCD detection is extremely sensitive and permits detailed study at high resolution of the effects of resonant FIR absorption by impurities (and free carriers) on individual PL lines. These effects result in an extremely complex and rich ODR spectrum, but the individual processes can be deconvolved by a simple fitting procedure. Studies concentrating on FIR absorption at the $1s$–$2p_+$ transition energy of neutral donors in GaAs/AlGaAs quantum wells show that at the lowest FIR power, the ODR spectrum does not result from carrier heating.

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