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OPTICALLY DETECTED IMPURITY D^0 AND D^- TRANSITIONS IN GaAs QUANTUM WELLS

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Abstract—We have carried out a far-infrared magnet-optical study on shallow donor states confined in GaAs quantum wells (QWs), applying a recently developed optical detection technique. We have observed, in addition to cyclotron resonance, the $1s \rightarrow 2p_+$ transition of neutral donors (D^0), and singlet and triplet transitions of negative donor ions (D^-); the latter observation verifies the existence of D^- ions in well-only doped QWs under optical pumping. This is the first observation of optically detected impurity resonances in confined systems and demonstrates the power and utility of this technique for such studies.

Over the past decade experimental studies of shallow donor states in semiconductor quantum wells (QWs) have exhibited a wealth of interesting physical phenomena[1]. The effects of confinement, magnetic fields and screening on energy levels of bound electrons in neutral donors (D^0) and negative donors (D^-) have been extensively studied, especially by far-infrared (FIR) magneto-spectroscopy. Shallow donor states in the presence of optical pumping, however, have not been well investigated. Under optical pumping conditions it is possible to obtain important insights into carrier and exciton dynamics, interaction between donors and photo-created electron-hole pairs, and the internal structure of excitonic complexes such as neutral-donor-bound excitons (D^0X) and negative-donor-bound excitons (D^-X).

Very recently a new type of optical detection technique has been developed by several groups for investigating FIR cyclotron resonance (CR) in semiconductors[2–5]. With this technique FIR resonance is detected through the change in the intensity of photoluminescence (PL) while the magnetic field is swept, rather than measuring FIR absorption directly. Remarkable sensitivity in comparison with conventional FIR methods has been demonstrated in studies of CR in several materials and structures. However, most of this work has concentrated on CR, and there has been only one detailed report of impurity studies [5]. In addition, despite the utility of this new detection scheme, few details of underlying mechanisms are understood. Hence, studies that elucidate the mechanisms and increase the range of applicability of this technique are of interest and importance.

In this paper we present results of a study of shallow donor states confined in GaAs QWs obtained with this optical detection technique. We have developed an apparatus that employs visible and FIR lasers, modulation of the FIR laser, and synchronous detection of changes in the near IR PL. We have observed, in addition to FIR CR, the $1s \rightarrow 2p_+$ transition of D^0, and singlet and triplet transitions of D^- ions. It should be noted that, since only the wells are doped with donors, this work represents a method for detecting D^- ion transitions in confined systems in which D^- *ions are optically created.* To emphasize its generality we call this technique "optically detected resonances" (ODR), instead of the conventionally used term, ODCR. ODR encompasses CR, impurity resonances, and also the possibility of intersubband and intra-excitonic resonances. The present results demonstrate the power and utility of ODR for investigating shallow impurity states in semiconductor nanostructures.

In the present experiment the sample was mounted in the Faraday geometry in a FIR lightpipe at the center of a 9-T super-conducting magnet cooled to 4.2 K. PL was excited with the 6328-Å line of a He-Ne laser via a 600-μm diameter optical fiber. The signals were collected with a second 600-μm fiber, and analyzed with a 0.25-m single grating spectrometer. The efficiencies of the FIR absorption and of the excitation and collection were maximized with a configuration in which the optical fibers were placed below the sample and the FIR radiation incident from above (see Fig. 1). A CO_2-pumped FIR laser was used to generate FIR radiation of wavelengths 393.6, 163.0, 118.8 and 96.5 μm. A personal computer was used to record simultaneously the magnetic field values and the detected changes in the PL, and to step the monochromator to follow the center of the desired PL peak as it shifted with magnetic field. With this system we were able to detect changes as small as 0.1% in the PL signal.

The sample used in this study was an MBE-grown $GaAs/Al_{0.3}Ga_{0.7}As$ multiple quantum well (MQW) structure doped in the center $\frac{1}{3}$ of the well with Si donors at $1 \times 10^{16}\,cm^{-3}$. The structure consists of six repetitions of a 210-Å well and 125-Å barriers. The PL [6] and FIR magneto-absorption[7] have been previously well studied.

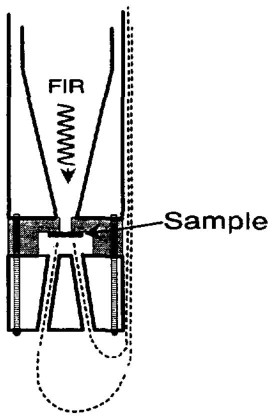

Fig. 1. Schematic diagram of the FIR light cone, sample, and optical fiber arrangement used for ODR.

In Fig. 2 we show the PL spectrum [in (a)] as well as the changes in the luminescence intensity induced by the FIR resonances at a wavelength of 118.8 μm [in (b)]. The PL peak at 1529.5 meV is due to free exciton recombination (e_1h_1), and the peak at 1527.7 meV is associated with donor recombination, dominated by neutral-donor-bound excitons[6]. The change (ΔI) in PL shown in Fig. 2b is associated with CR; the field was kept constant (at the value corresponding to CR) as the detected energy was varied. The free exciton PL intensity *increases* due to the CR, whereas the bound exciton PL intensity *decreases*. These changes in PL are explained through a resonant electron heating effect. As the effective temperature of the electrons is increased by CR, the bound excitons dissociate into free excitons and donors. This process increases the free exciton population; hence the free exciton PL intensity increases and the bound exciton PL decreases. For magnetic field values corresponding to impurity resonances, similar effects are observed. In such cases a two-step "photothermal" process[8], in which the bound electrons in the excited states of donors (after absorbing FIR light) are thermally ejected into the conduction band with a high probability (about unity for GaAs at the experimental temperatures in the present work), ionizes donors. This process thus decreases the number density of neutral and negatively-charged donors in a steady state, and also allows the interaction between the liberated electrons and the surroundings, both of which contribute to increase (decrease) the intensity of free (bound) exciton PL.

Two scans of the change in PL intensity (ΔI) as a function of magnetic field (ODR signal) for a FIR laser line of 118.8 μm are presented in Fig. 3. The upper trace shows the *positive* change (i.e. increase) in the intensity of the free exciton luminescence, whereas the lower trace shows the *negative* change (i.e. decrease) in the intensity of the bound exciton luminescence. For both scans four different FIR resonances are clearly seen with an excellent signal-to-noise ratio. Accompanying the electron CR [feature (c)] observed at 6.2 T are three donor-related resonances: (a) the $1s \to 2p_+$ transition of the neutral donor (D^0) confined in the GaAs QWs, (b) the singlet transition of negative donor ions (D^-), and (d) a triplet transition of D^-. These are the first data to show optically

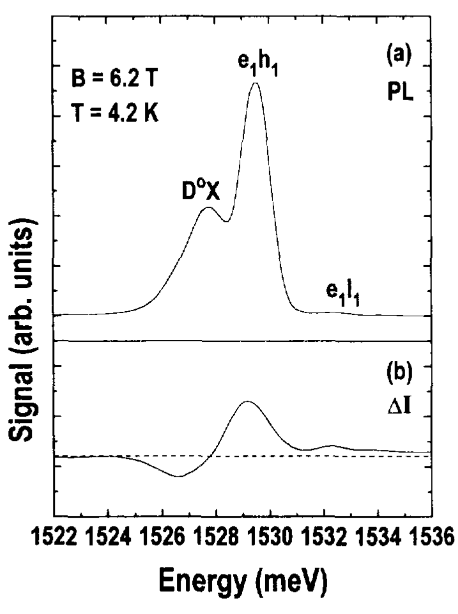

Fig. 2. (a) The PL spectrum, and (b) the change (ΔI) in PL intensity due to FIR absorption observed at 6.2 T, which corresponds to electron CR at 118.8 μm.

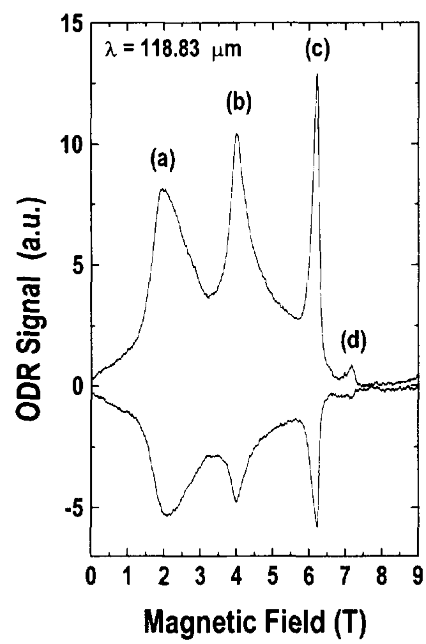

Fig. 3. Two field scans at a FIR wavelength of 118.8 μm at 4.2 K with a pumping power of 0.68 W cm^{-2}. The ODR signal represents the change in the free exciton PL (upper trace) and the bound exciton PL (lower trace). The observed features are: (a) the $1s \to 2p_+$ transition of D^0, (b) the singlet transition of D^-, (c) electron CR, and (d) a triplet transition of D^-.

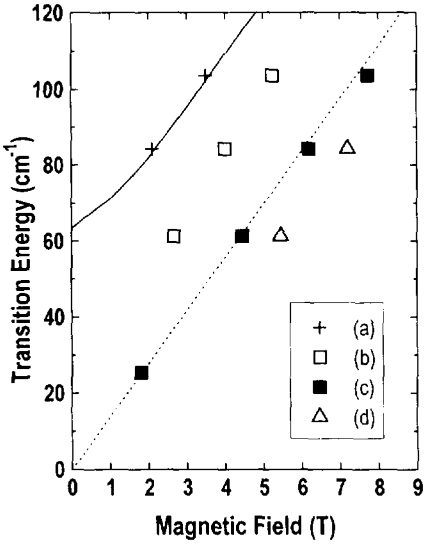

Fig. 4. Experimentally observed peak positions vs field, together with theoretical curve (solid line) for feature (a) from Ref. [9] and a straight dotted line that corresponds to $0.069m_0$ for CR.

detected impurity resonances in confined systems to the best of our knowledge.

The resonance positions for the features observed with four FIR laser lines are plotted against magnetic field in Fig. 4. The identification of these features was made through comparison with earlier transmission and photo-conductivity data[7,11,12,14] on similar well-center-doped MQW structures with the same well-width. The $1s \rightarrow 2p_+$ transition of D^0, feature (a), has been extensively studied and is well understood [1,7]; its resonance position is in excellent agreement with variational calculations for this well-width[9] (solid curve in Fig. 4). Feature (b) is due to the singlet transition of D^- (first reported by Huant *et al.*[10] and for which the transition energy vs field is now well established[11]). The origin of the small feature (d), observed at 7.2 T for 118.8 μm in Fig. 3, is somewhat controversial. Both a triplet transition of D^- and the $2p_- \rightarrow 2s$ transition of D^0 have been suggested as possible interpretations[12,13]. However, recently both of these features have been simultaneously observed in a single sample and identified via a detailed temperature dependence study[14]. Based on these results, we assign the weak line observed in the present work to the low-frequency triplet transition. For available FIR laser lines with shorter wavelengths (higher energies), the resonant field for feature (d) shifts above 9 T (the maximum field available with the present magnet). In the cases of the longest FIR wavelengths, the photon energies are lower than the transition energies for (a) and (b) at zero field, and hence features (a) and (b) do not appear in the spectra.

It is striking that a D^- singlet line with a larger intensity than the D^0 line is observed in this well-only doped sample. Generally, with conventional FIR methods, D^- lines are observed only when *both* the well and barrier layers are doped, such that there are more electrons available than donors in the well. Under these conditions, D^- ion transitions are observed at low temperatures[10–12]. However, in the present situation there are no excess electrons in the dark, and hence no D^- ions can exist in the absence of optical and thermal excitations: D^- ions observed in our QWs are thus created by the optical pumping. Furthermore, the appearance of the D^- triplet transition implies that the effective temperature in the system is relatively high. In transmission or photoconductivity experiments[14] we observe this line only above 15 K; the triplet ground state, which is higher in energy than the singlet ground state, is populated only at higher temperature.

The coexistence of several features in the ODR spectra suggests a complicated steady-state for doped QWs under optical pumping. In such a state some donors are positively ionized (D^+), others bind two electrons (either singlet $D^-_{\uparrow\downarrow}$ or triplet $D^-_{\uparrow\uparrow}$), and still others remain neutral (D^0); all coexist with free electrons, free holes, and excitons that are either free or bound to any of the above-mentioned donors. The realization of this situation and the detailed information about the populations of these states and complexes demonstrates the power of ODR. The experimental fact that we observe the well-known $1s \rightarrow 2p_+$ hydrogenic transition in D^0 without any energy shift means that we are *not* probing the internal structure of donor-bound excitons, whose energy structure should be completely different. Since the number density of photo-created electron-hole pairs is much lower than that of donors, most donors do not form excitonic complexes, remaining isolated hydrogenic neutral donors. Efforts to create an experimental situation in which internal transitions of excitonic complexes are observable, and to understand microscopic mechanism which leads to the optical detection of impurity resonances are in progress[15].

In summary, we have observed the $1s \rightarrow 2p_+$ transition of D^0, singlet and triplet transitions of D^-, as well as CR, in well-center-doped GaAs QWs, employing the recently developed optical detection technique, ODR. We have shown that the applicability of this new technique is not limited to CR but to many types of FIR resonances. As an example of its power, we have discovered the existence of D^- ions in doped QWs under optical excitation. Further possibilities of detecting intersubband resonance and intra-excitonic resonance with this technique are being tested. In addition, preliminary results obtained by using reflectivity and photoconductivity (instead of PL) for ODR appear promising for studies of materials and structures that luminesce poorly due to strong non-radiative recombination[15].

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