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Optically detected far-infrared resonances in doped GaAs quantum wells

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We have observed a variety of far-infrared resonances, including the transition from the ground state to the first excited state in neutral donors, and singlet and triplet transitions of negative donor ions (D^-), as well as electron-cyclotron resonance, in well-center-doped GaAs quantum wells, employing a recently developed optical detection technique. The power of this technique for studying impurity states in confined systems is clearly revealed. Results provide evidence for the existence of D^- centers under optical excitation in multiple-quantum-well structures doped only in the wells.

During the past few years a new type of optical detection technique has been developed by several groups for investigating far-infrared (FIR) cyclotron resonance (CR) in semiconductors.[1–7] Because of some significant advantages over conventional CR methods, there has been considerable interest in this novel technique. One of the advantages, apart from high sensitivity and resolution, lies in the possibility of selecting spectrally a specific photoluminescence (PL) feature, among various band-edge features, as the detection "channel." Since the typical PL spectrum from semiconductors involves free excitons, bound excitons, and band-to-impurity, impurity-to-band, or impurity-impurity (donor-acceptor) recombination lines, it should be possible to use the specificity of the FIR spectrum to obtain information about recombination mechanisms and the interactions that give rise to the various lines. Also, for new materials or new structures, the combination of FIR resonance and PL should permit the unambiguous identification of particular features in the PL spectrum.

In this detection scheme, FIR (or microwave) resonance is detected through the change in the intensity of PL while the magnetic field is swept, rather than measuring FIR (or microwave) absorption directly. The feasibility of such a detection scheme was shown by Baranov *et al.*[8] and subsequently by Romestain and Weisbuch[9] in the microwave range, and recently this general technique has been extended to the FIR region of the spectrum.[1–7] Wright and co-workers[1,2] have observed, in high-purity GaAs epilayers, the electron CR spin-doublet splitting, electron polaron effects, and the light-hole quantum CR, as well as the donor $1s \rightarrow 2p_+$ transitions, demonstrating the remarkable sensitivity of this technique. It should be noted that the donor $1s \rightarrow 2p_+$ transitions in most semiconductors occur in the FIR, and thus cannot be detected by microwave methods. Subsequently, several other groups have used this technique to study CR in other materials and structures.[3–7] Most of this work has concentrated on CR, and there has been only one detailed report of impurity studies.[7] In addition, the application of this technique has not been well explored in low-dimensional systems to date. Finally, despite the utility of this 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 the discussion that follows we present results of such a study. 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. Applying this technique to well-center-doped GaAs quantum wells (QW's), we have observed, in addition to FIR CR, the $1s \rightarrow 2p_+$ transition of neutral donors (D^0), and singlet and triplet transitions of negative donor ions (D^-). It should be noted that, since only the wells are doped with donors, this work represents an excellent 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 intraexcitonic resonances. The present results demonstrate the power and utility of ODR for investigating shallow impurity states in semiconductor nanostructures.

Figure 1(a) is a schematic of the experimental apparatus used for the observation of ODR. The sample was mounted in the Faraday geometry in a FIR light pipe at the center of a 9-T superconducting magnet cooled to 4.2 K. PL was excited with the 6328-Å line of a He-Ne laser via a 600-μm-diam optical fiber. The signals were collected with a second 600-μm fiber, and analyzed with a 0.25-m single-grating spectrometer/Si-diode detector combination. The excitation-collection was optimized by orienting the axis of each fiber 8° from the normal to the sample surface. 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(b)]. A CO_2-pumped FIR laser was used to generate FIR radiation of wavelengths 393.6, 163.0, 118.8, and 96.5 μm. The FIR laser power supply was operated in the chopped mode with the lock-in amplifier referenced to this chopped signal. A dedicated 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 a molecular-beam-epitaxy-grown GaAs/$Al_{0.3}Ga_{0.7}As$ multiple-quantum-well (MQW) structure doped in the center third of the well with

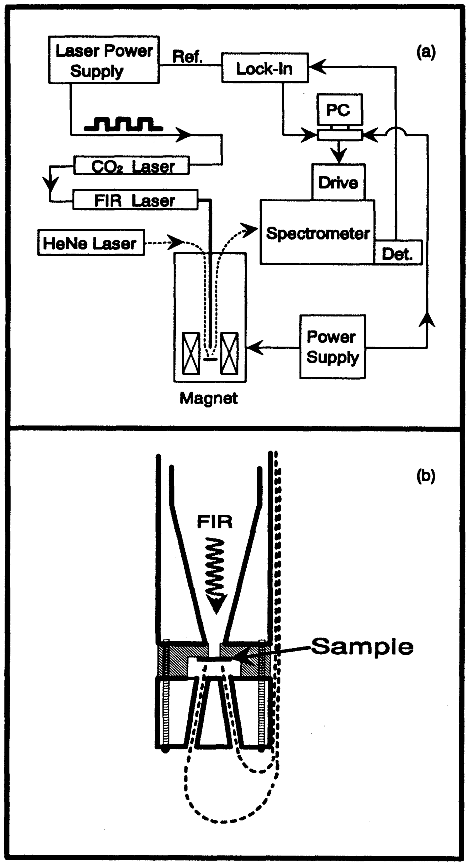

FIG. 1. (a) Schematic diagram of the experimental setup for ODR. (b) Diagram of the FIR light cone, sample, and optical fiber arrangement.

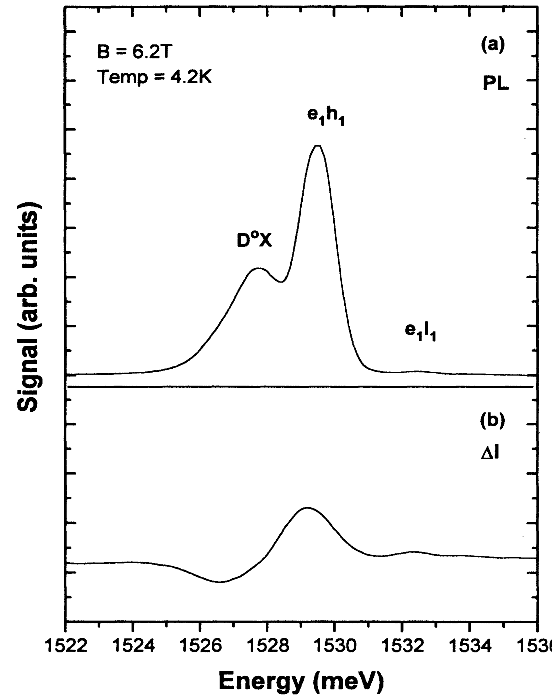

FIG. 2. (a) The PL spectrum, and (b) the change (ΔI) in PL intensity due to FIR absorption observed at 6.2 T, which corresponds to electron cyclotron resonance at the laser wavelength of 118.8 μm. The magnitude of ΔI at the e_1h_1 peak corresponds approximately to 1% of the total PL intensity.

Si donors ($N_D = 1\times10^{16}$ cm^{-3}). The MQW structure consists of six repetitions of a 210-Å well and 125-Å barriers. The PL from this sample has been previously well studied.[10]

In Fig. 2 we show the PL spectrum as well as the changes in the luminescence intensity induced by the FIR resonances at a wavelength of 118.8 μm and a magnetic field of 6.2 T. The PL peak at 1529.5 meV is due to free-exciton recombination involving the ground-state heavy-hole subband (e_1h_1), and the peak at 1527.7 meV is associated with donor recombination, dominated by neutral-donor bound excitons;[10] the weak feature at 1532.6 meV is identified as the free-exciton recombination due to the ground-state light-hole subband (e_1l_1). Figure 2(b) shows the change (ΔI) in PL associated with CR; the field was kept constant (at the value corresponding to CR) as the detected energy was varied. The difference spectrum in Fig. 2(b) shows that 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,[11] 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.

Four scans of the change in the free-exciton luminescence intensity (ΔI) as a function of magnetic field (ODR signal) for FIR lines at 393.6, 163.0, 118.8, and 96.5 μm are presented in Fig. 3. At $\lambda = 118.8$ μm, four resonances are easily seen. Accompanying the electron CR [feature (c)] are three donor-related resonances. Feature (a) is the $1s \rightarrow 2p_+$ transition of the neutral donor (D^0), and feature (b) [(d)] is the singlet (triplet) transition of the negatively ionized donor (D^-). 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 (see inset to Fig. 3).

The identification of the observed impurity lines was made through comparison with earlier transmission and photoconductivity data[12,15,16,18] on similar well-center-doped MQW structures with the same well width. The $1s \rightarrow 2p_+$

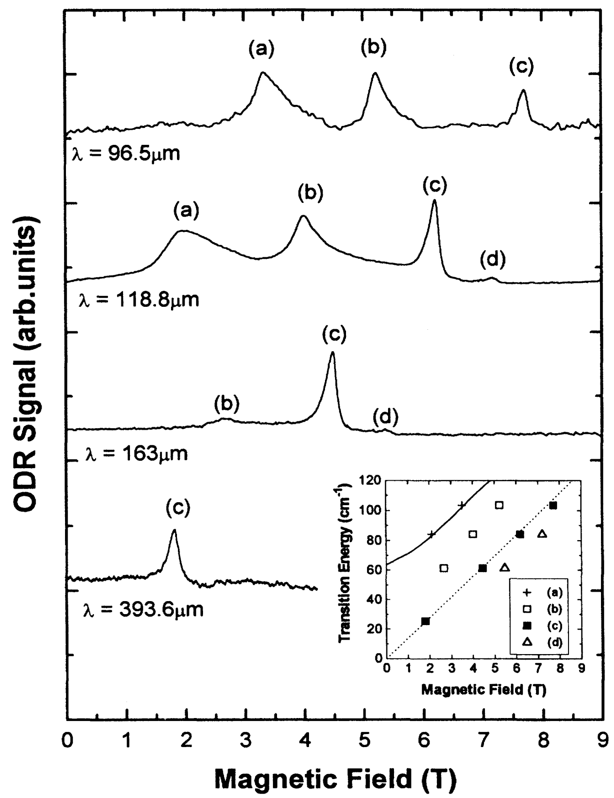

FIG. 3. Four field scans at FIR wavelengths of 96.5, 118.8, 163, and 393.6 μm at 4.2 K with a pumping power of 0.68 W/cm^2. The ODR signal represents the change in the free-exciton PL. The observed features are (a) the $1s \rightarrow 2p_+$ transition of neutral donors (D^0), (b) the singlet transition of negative donor ions (D^-), (c) electron CR, and (d) the triplet transition of D^-. The inset shows experimentally observed peak positions vs field, together with a theoretical curve (solid line) for the feature (a) from Ref. 13 and a straight dotted line that corresponds to $0.069m_0$ for CR.

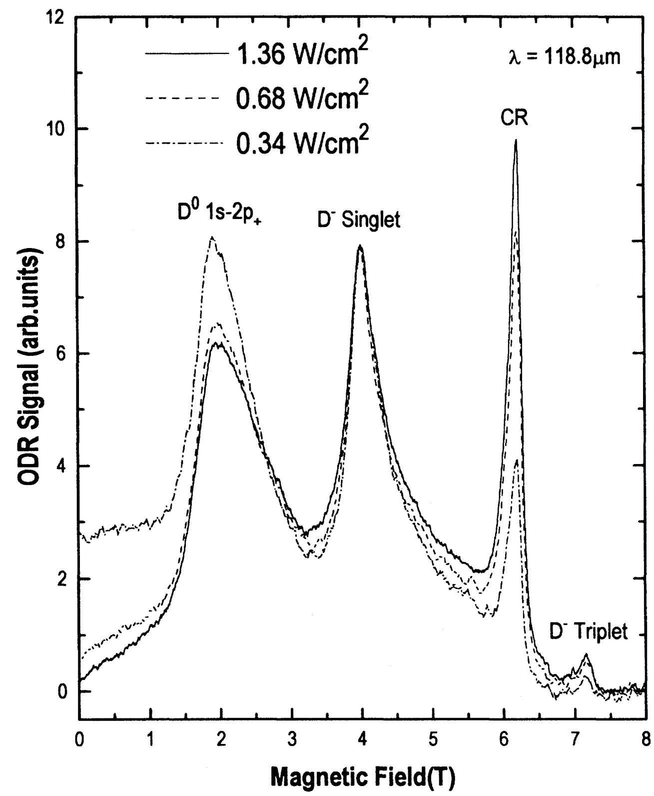

FIG. 4. The He-Ne laser pumping power dependence of ODR spectra at a FIR wavelength of 118.8 μm. The monochromator was set to detect the free-exciton PL. The spectra are normalized in such a way that the signal intensity of the D^- singlet line becomes equal for all three spectra.

transition of D^0, feature (a) in Fig. 3, has been studied and is well understood;[12] its resonance position is in excellent agreement with variational calculations for this well width[13] (see inset to Fig. 3). Feature (b) in Fig. 3 is due to the singlet transition of D^- (identified by Huant *et al.*[14] and for which the transition energy vs field is now well established[15]). The origin of the small feature (d), observed at 7.2 T for 118.8 μm, is somewhat controversial. Both a triplet transition of D^- and the $2p_- \rightarrow 2s$ transition of D^0 have been suggested as possible interpretations.[16,17] However, recently both of these features have been simultaneously observed in a single sample and identified via a detailed temperature-dependence study.[18] Based on these results, we assign the weak line observed in the present work to the low-frequency triplet transition.

It is striking that a D^- singlet line with a comparable intensity with 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.[14–16] 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 QW's are thus created by the optical pumping. The coexistence of several features in the ODR spectra suggests a complicated but interesting steady state. In such a state some donors are 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 fact that the D^- singlet line has an intensity comparable to those of both the D^0 and CR lines suggests that the number density of D^- ions is comparable to that of D^0 and of free electrons in the conduction band. Furthermore, the appearance of the D^- triplet transition implies that the effective temperature in the system is relatively high. In transmission or photoconductivity experiments,[18] 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 realization of this situation under optical excitation and the detailed information about the populations of these states and the electron temperature demonstrates the power of ODR.

In Fig. 4 ODR signals for the free-exciton line are displayed as functions of magnetic field for three different pumping powers (0.34, 0.68, and 1.36 W/cm^2) of the 6328-Å line. Here, all the spectra are scaled so as to make the intensities of the D^- singlet lines equal for all three spectra. In this way we can focus our attention on relative intensities rather than absolute intensities; absolute intensities cannot be directly compared because of the power dependence of PL intensity itself. Since different pumping powers create different steady states with different population ratios for different types of donor states (i.e., D^+, $D^-_{\uparrow\downarrow}$, $D^-_{\uparrow\uparrow}$, and D^0), we observe an interesting evolution of relative intensities for different features. The relative intensity of CR increases with pumping power continuously and monotonically. An interesting feature observed for the 0.34- and 0.68-W/cm^{-2} spectra is the low-field tail, which we assign to the zero-field donor-ionization transition since its energy for this well width (210 Å) coincides with the photon energy (10.4 meV) of the FIR radiation. This is an allowed transition at zero field, whose oscillator strength decreases with field, vanishing in the high-field limit. To obtain a rough idea of the occupancy of the neutral donor ground state, we integrate the ODR curves from $B=0$ to 3.5 T to obtain the area under the curves. The area decreases continuously as pump power increases. Thus we attribute the *increase* of CR and decrease of the zero-field signal and integrated area up to 3.5 T as due to an increase in the effective temperature of the system with pump power. A detailed statistical analysis for at least five energy levels is necessary for a quantitative clarification of the observed evolution (much like the analysis employed in Ref. 16 for a three-level system) and further investigations are in progress.[19]

In summary, we have applied the recently developed technique ODR to well-center-doped GaAs QW's and observed several absorption lines: the $1s \rightarrow 2p_+$ transition of neutral donors (D^0), singlet and triplet transitions of negative donor ions (D^-), as well as electron CR. We have shown that the applicability of this technique is not limited only 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 QW's under optical excitation. Further possibilities of detecting intersubband resonance and intraexcitonic resonance with this technique are being tested. In addition, our 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 nonradiative recombination.[19]

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