Compensation effects on optically pumped intersubband absorption in $GaAs/Al_xGa_{1-x}As$ multiple-quantum-well structures

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Measurements of intersubband transitions and cyclotron resonance (CR) have been carried out on lightly donor-doped $GaAs/Al_{0.3}Ga_{0.7}As$ multiple-quantum-well structures. Absorption coefficients of CR and intersubband resonances combined with calculations of coupling efficiency, give an oscillator strength of about 1. Measurements of CR absorption versus pump intensity show clear saturation at high intensities. Results demonstrate that the observed intersubband transitions are due to free electrons and that the large photogenerated excess free-electron density is due to compensation by acceptors.

Recent experimental studies of intersubband optical transitions in GaAs/Al-Ga-As multiple-quantum-well (MQW) structures have made use of a sensitive pump and probe technique in which a light-emitting diode (LED) or laser with photon energy greater than the gap of the wide-band-gap semiconductor served as the pump source while the intersubband absorption was detected in the infrared (ir).[1,2] Results for nominally undoped GaAs/Al-Ga-As MQW's were attributed to the photogenerated heavy-hole transitions between excitonic states associated with the first and second conduction subband.[1] From the total integrated absorptance and laser power, an extremely large oscillator strength for this transition was inferred ($f=40$ for exciton lifetime $\tau=2$ nsec). It was argued that this oscillator strength was in fact an "effective excitonic oscillator strength" which was greatly enhanced in QW structures.[3-5]

In this paper we present a detailed study of photoinduced intersubband absorption and *simultaneous* measurements of the photoinduced free-carrier density via cyclotron resonance (CR) in donor-doped MQW structures with metallic grating couplers. These measurements, combined with a numerical calculation of the grating coupling efficiency, allow the determination of the free-carrier intersubband oscillator strength. Results show that extremely large excess free-electron densities are created under optical pumping conditions at temperatures above 30 K, indicating an important modification of the optically pumped spectroscopy by compensation. This assertion is confirmed by the pump intensity dependence of cyclotron resonance in similar samples without grating couplers. We also show that free-carrier and excitonic intersubband oscillator strengths are comparable.

Intersubband measurements were performed on two $GaAs/Al_{0.3}Ga_{0.7}As$ MQW samples: (1) nominal well width of 240 Å, barrier width of 240 Å, 30 periods, and Si dopant density 4×10^{16} cm^{-3} over the central 80-Å region of each barrier; (2) nominal well width of 320 Å, barrier width of 125 Å, 12 periods, and Si dopant density 1×10^{16} cm^{-3} over the "top $\frac{1}{3}$" of each well.[6] Metallic grating couplers were deposited on the sample surfaces by conventional photolithography and lift-off techniques. The grating couples the transverse electric-field component of the normally incident infrared radiation into the longitudinal polarization (the direction of confinement). An *in situ* red-light-emitting diode placed close to the sample surface was square-wave modulated at audio frequency in order to generate excess free carriers in the wells. The ir transmitted signal coherent with the LED modulation was detected by a Ga:Ge photoconductive detector at 4.2 K, amplified and coherently demodulated by a lock-in amplifier. The dc output voltage from the amplifier was then digitized and Fourier transformed by a microcomputer into a frequency spectrum, which is the difference spectrum between LED-on and LED-off.

Figure 1 shows the differential spectrum taken from the 240-Å-well-width sample at a magnetic field of 8 T and sample temperature $\simeq 25$ K. Feature ISB, which is independent of magnetic field, is the intersubband transition. The cyclotron resonance (CR), which shifts with magnetic field at about 13.5 cm^{-1}/T, is due to the residual in-plane component of electric field in the vicinity of the grating. The intensities of both ISB and CR increase when the temperature is increased due to the generation of free electrons through thermal ionization.[7] Table I is the summary of results. The calculations for the subband energies were made with a one-dimensional square-well model with barrier height of 230 meV and well width determined from photoluminescences (PL) and reflectivity measurements.[8] The subband transition energies from ir measurements show lower values (outside the overall experimental error) than calculated. This is likely due to final-state interactions.[9] In general, the intersubband resonance energy is approximately given by $\tilde{\omega}_{21}^2$

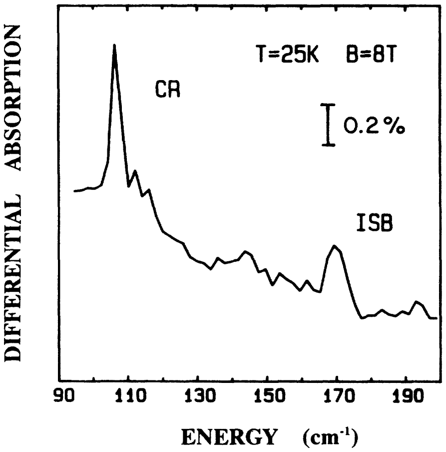

FIG. 1. Differential absorption spectrum of a 240-Å-wide MQW sample at $B=8$ T and $T\approx 25$ K. CR is the cyclotron resonance and ISB is the intersubband resonance.

$=\omega_{21}^2(1+a-b)$, where ω_{21} is the subband separation, a is a correction for depolarization effects, and b is the exciton-like final-state effect. The depolarization effect is important at high carrier concentration. (For a two-dimensional density of 10^9 cm^{-2} the shift in frequency due to depolarization is about 1%.) At low electron concentrations the final-state interaction is more important than the depolarization effect, and the resonance energy becomes slightly smaller than the subband separation.

In order to extract the oscillator strength from experimental data, we need to determine the fraction of in-plane electric field coupled into the z direction and the photogenerated electron density. The coupling efficiency of a grating, η, is defined as the ratio of $\langle E_z^2\rangle$ to $\langle E_0^2\rangle$ (spatial average over the MQW structure), where E_z is the z component of electric field underneath the grating, and E_0 is the incident in-plane electric field.[10] The intersubband absorption coefficient, α, is related to the efficiency η by

$$\alpha=\frac{4\pi}{c}\epsilon^{3/2}\frac{\mathrm{Re}\sigma_z}{|\epsilon_z|^2}\eta\,, \tag{1}$$

where ϵ_z (σ_z) is the QW dielectric function (conductivity) from the anisotropic plasma model,[11] and ϵ is the dielectric constant of GaAs. The oscillator strength f is defined as

$$f\equiv\frac{2m^*\tilde{\omega}_{21}}{\hbar}|\langle F_2|z|F_1\rangle|^2\,, \tag{2}$$

where m^* is the electron effective mass, $\tilde{\omega}_{21}$ is the subband resonance frequency, and $\langle F_2|z|F_1\rangle$ is the z-component dipole matrix element between the quantum-well envelope functions, F_1 and F_2, for the first and second subbands, respectively. $\mathrm{Re}\sigma_z$ contains the oscillator strength, the intersubband resonance frequency, the intersubband absorption linewidth, and the plasma frequency $\omega_p^2=4\pi N_e e^2/m^*L$ (L is the well width). Therefore, the oscillator strength can be obtained from Eq. (1) with the experimentally measured α, the electron density N_e, and η for a particular sample geometry. Results, shown in Table I, are in good agreement with the theoretical value of 0.96 obtained directly from Eq. (2) with $F_1=(2/L)^{1/2}\times\sin(\pi z/L)$, $F_2=(2/L)^{1/2}\sin(2\pi z/L)$, and $m^*=0.068m_0$ (m_0 is the free-electron mass).

The fact that only CR and intersubband absorption lines are observed, and no signs of exciton internal transitions, shows that the intersubband transitions are due to free electrons. If each photon creates one electron-hole pair (quantum efficiency equals one) with a recombination time, τ, of 1 nsec, the photoinduced carrier density estimated from $N_e\leq g\tau$ with LED photon flux $\simeq 2\times 10^{14}$ cm^{-2}sec^{-1} (60 μW cm^{-2}) is of the order of 2×10^5 cm^{-2}. This is four orders of magnitude smaller than the number density per well obtained from the *in situ* CR measurement ($N_e\sim 10^9$ cm^{-2}). We suggest that the large excess carrier density is due to compensating acceptors combined with a very long electron-acceptor lifetime (of the order of μsec). This is possibly due to spatial separation of the electrons from the acceptors which could be located at the interfaces or $Al_{0.3}Ga_{0.7}As$ layers.[12] These acceptors are unintentionally incorporated into the sample during growth. The existence of residual carbon acceptors has been confirmed by PL experiments.[8] If such compensating acceptors are responsible for the excess carrier density, the optically induced CR should saturate at high pump intensity.

To test this idea, we have performed CR experiments on two separate MQW samples (labeled 3 and 4, respectively) grown under similar conditions to those for samples 1 and 2. Sample 3 (30 wells) and sample 4 (20 wells) have the same nominal well width of 210 Å, barrier width of 125 Å, and the same Si dopant density of 1×10^{16} cm^{-3} over the central $\frac{1}{3}$ of each well. These two samples cover a wide range of compensation. From PL measurements, sample 3 is known to be significantly more compensated than sample 4. A 35-mW He-Ne laser was used as a pump source, and the laser light was directed onto the sample surface by an optical fiber. Figure 2 shows CR spectra taken from sample 3 at various laser intensities on the sample surface. The sample was tilted 20° with respect to the direction of the magnetic field in order to separate two-dimensional CR from three-dimensional CR. Saturation is evident from a plot of absorption intensity as a function of laser power [Fig. 3(a)]. Sample 3 has

TABLE I. Summary of results.

	Well width (Å) (PL and reflectivity)	ω_{21} (cm^{-1}) (calculated)	$\tilde{\omega}_{21}$ (cm^{-1}) (ISB Expt.)	η (%)	f
Sample 1	240 ± 5	181 ± 7	170 ± 3	15.5	1.0 ± 0.3
Sample 2	320 ± 5	110 ± 3	99 ± 3	26.5	1.1 ± 0.3

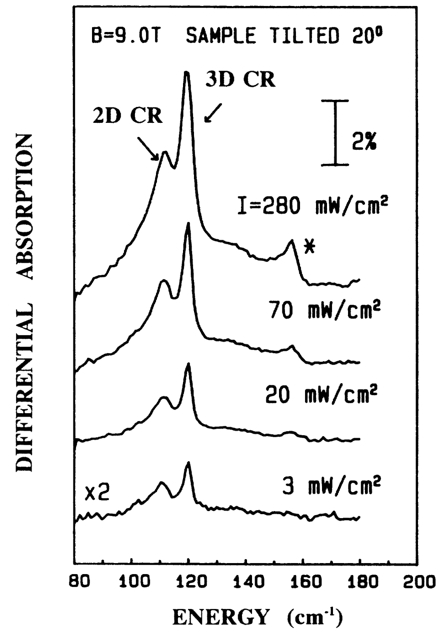

FIG. 2. Cyclotron resonance spectra taken from sample 3 at $B=9.0$ T, $T\approx 50$ K for various laser pump intensities. The normal to the sample surface was tilted 20° away from the magnetic-field direction in order to separate two-dimensional CR from three-dimensional CR. The feature labeled * at 156 cm^{-1} is the characteristic D line of the bulk carbon acceptors in GaAs.

higher absorption strengths than sample 4, indicating a higher degree of compensation in this sample. The relation between absorption strength I_a and light intensity I_p, usually fitted to a power law of the form $I_a \propto I_p^{\gamma}$, is useful in determining the dominant recombination process.[13] Least-squares fits of $\log(I_a)$ vs $\log(I_p)$ at low pump intensities yields values of $\gamma=0.50\pm0.04$ and 0.56 ± 0.05, for samples 3 and 4, respectively [Fig. 3(b)]. This shows that recombination in both samples is dominated by a bimolecular-type process in which carriers are liberated from trapping centers under light illumination. The differential CR absorption intensity is proportional to the change of free-carrier density ΔN_e between light on and off. Since the free electrons must come from thermal ionization of neutral donors under continuous pumping, ΔN_e should be equal to the change in neutral donor density ΔN_D. In the dark and at low temperatures these donors are ionized by compensating acceptors. Therefore the transition rate (recombination rate, $1/\tau_{D\text{-}A}$) from neutral donor states to neutral acceptor states is proportional to the number of neutral acceptors ΔN_A created by the light. At the temperatures of the experiments there is essentially no *thermal* ionization of acceptors. Since $\Delta N_D \propto I_p \tau_{D\text{-}A} \propto I_p/\Delta N_A$ and $\Delta N_D=\Delta N_A$, we arrive at the relation $I_A \propto \Delta N_D \propto I_p^{0.5}$, in agreement with experimental observation. (Note: since $\Delta N_e=\Delta N_D$, this argument is also appropriate for free-electron–neutral-acceptor recombination.) This confirms the impurity-related free-carrier generation. The exciton recombination is a monomolecular process in which $\gamma\simeq 1$ because the exciton lifetime is basically independent of pump intensity.

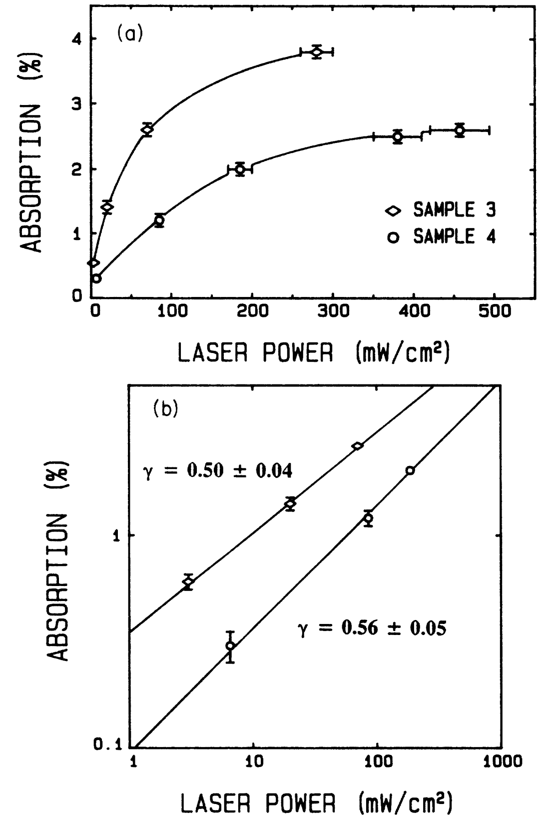

FIG. 3. Dependence of cyclotron resonance absorption on laser power I_p for sample 3 and sample 4. (a) Saturation of absorption intensity I_a, and (b) plot of $\langle I_a\rangle$ vs $\langle I_p\rangle$ on a doubly logarithmic scale. γ is obtained via a least-squares fit of the expression $\log(I_a)=\gamma\log(I_p)+C$ to the first three data points. C is a constant.

Finally, we consider the free-exciton intersubband oscillator strength, in comparison to the free-electron intersubband oscillator strength, in order to determine if the exciton mechanism[1] could provide a possible explanation. In a quantum well, the free-exciton envelope wave function can be written as

$$\psi=\frac{e^{i\mathbf{k}\cdot\mathbf{R}}}{\sqrt{A}}\phi^{2\mathrm{D}}(\rho)F(z_e)F(z_h)\,, \tag{3}$$

where $\phi^{2\mathrm{D}}(\rho)$ is the two-dimensional exciton envelope function with $\rho=\rho_e-\rho_h$, $\mathbf{R}$ is the center-of-mass coordinate in the plane of the well, A is the area of the sample, and $F(z_e)$ and $F(z_h)$ are the quantum well envelope functions of electron and hole, respectively. In a confined system, the oscillator strength for excitonic transitions across the gap is estimated to be 13 times that of the bulk (for well width of 50 Å) (Ref. 3) because the overlap of the electron-hole envelope function is greatly enhanced.[3-5] The strength of the *intersubband* transitions is determined in general by the matrix element of the dipole operator in the z direction,

$$\langle\psi_2|z_e|\psi_1\rangle=\langle\phi_2(\rho)|\phi_1(\rho)\rangle\langle F_2(z_e)|z_e|F_1(z_e)\rangle\,, \tag{4}$$

where 1 and 2 denote the initial and final set of quantum numbers. For free carriers, $\phi_1(\rho)$ and $\phi_2(\rho)$ are identical plane-wave states. For excitons, $\phi_1(\rho)$ and $\phi_2(\rho)$ are the confined 1*s* hydrogenic envelope functions in the *x*-*y* plane associated with the first and the second subband, respectively. $\phi_1(\rho)$ and $\phi_2(\rho)$ are very similar; in fact, they are exactly the same in a strictly two-dimensional system.[14] Therefore $\langle \phi_2(\rho) | \phi_1(\rho) \rangle \cong 1$. Since the *z* part of the envelope functions for free carriers and excitons are also very similar, Eq. (4) implies that the strength of the excitonic intersubband transition and that of free-electron intersubband transition are essentially the same, and there is no "giant excitonic oscillator strength" associated with such transitions.

In summary, we have performed a series of far-infrared experiments to study the intersubband transitions in MQW structures with low doping densities and the effects of compensation on the optically pumped spectroscopy. Results show that compensation plays an important role, the photoinduced carrier density in the presence of compensating acceptors can be several orders of magnitude larger than that without compensation, and the observed intersubband transitions are due to free electrons. There is no substantial difference in oscillator strength between exciton and free-electron intersubband transitions.

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