Photoinduced Intersubband Absorption in Undoped Multi-Quantum-Well Structures

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We report the first observation of strong intersubband ($e1$-$e2$) absorption by photogenerated excitons in GaAs/GaAlAs undoped, multi-quantum-well structures. The origin of the transition is verified by polarization measurements, temperature dependence, and comparison with photoluminescence data. Its oscillator strength is found to be approximately 25 times stronger than that measured for bare electrons in modulation-doped quantum wells. We attribute it to an excitonic enhancement.

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The electronic levels of multi-quantum-well (MQW) structures, such as GaAs/GaAlAs, are usually studied by optical transitions between the conduction and valence bands.[1] Recently there have been several reports on infrared (ir) transitions between the electron subbands in doped MQW structures.[2-4] The observed absorption corresponds to the $e1$-$e2$ transition and is polarized parallel to the MQW growth direction (z axis). Its absorption strength is large, and it agrees with theoretical estimates of electron transitions in strictly one-dimensional quantum wells. Interest is drawn to this absorption as it could be utilized for ir detection, with a cutoff wavelength determined by the MQW parameters.

In this Letter we report the first observation of direct, ir intersubband photoinduced absorption (PA) by photogenerated free excitons, in an *undoped* GaAs/GaAlAs MQW structure. It is identified as the resonant transition from the ($e1$:hh1) to the ($e2$:hh1) exciton subband. The assignment is based on polarization measurements and on a comparison with the energies of the electron and hole subbands as measured by photoluminescence excitation (PLE). A consistent energy-level scheme is obtained and the model calculations provide the quantum-well parameters. The oscillator strength of the PA is found to be $\simeq 25$ times stronger than the corresponding absorption by bare electrons in doped quantum wells. Recent theoretical calculations of the oscillator strength of interband excitonic absorption in quasi-two-dimensional QW's (i.e., free motion in the x-y plane and confinement in the z direction) showed that it is greatly enhanced due to the large coherence area of the exciton.[5-7] Using the experimental oscillator strength we estimate the exciton coherence length to be ~ 5 exciton radii, or ~ 250 Å.

The GaAs/GaAlAs MQW sample was grown by a Perkin-Elmer 425B molecular-beam-epitaxy (MBE) system on an undoped, (001)-oriented GaAs substrate. The nominal parameters of the MQW structure are as follows: a set of 33 undoped GaAs quantum wells, 50 Å wide, separated by undoped $Al_{0.33}Ga_{0.67}As$ barriers, 100 Å wide. In all the experiments, the sample was excited by either a cw Ar^+-ion laser at 5145 Å, deep into the conduction band, or selectively into individual exciton levels by a cw dye laser. In order to accurately determine the laser intensity exciting the crystal, the power was measured in front of the Dewar and was then corrected for both window- and sample-surface reflectivity. The laser beam covered the whole sample face. For the ir absorption measurements we have fabricated a multipass waveguide by polishing two (011) faces at a 45° angle, allowing ir transmission as shown schematically in the inset of Fig. 1. The PA measure-

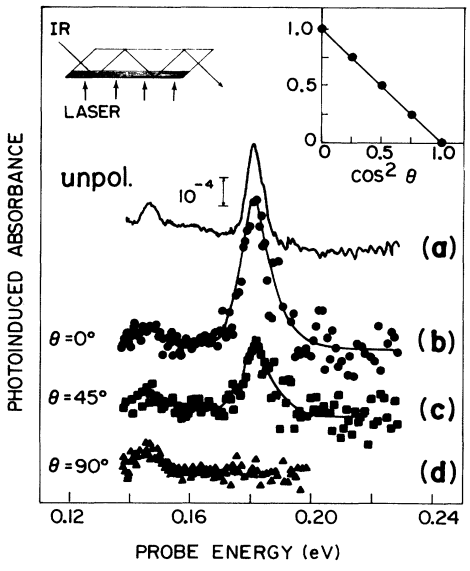

FIG. 1. The intersubband PA spectra of $GaAs/Al_{0.33}Ga_{0.67}As$ MQW at 90 K for various angles θ of the ir electric field with respect to the MQW growth axis. Excitation is at 2.409 eV with 0.1 W/cm^2. (a) Unpolarized light, (b) $\theta=0$, (c) $\theta=45°$, and (d) $\theta=90°$. Insets: The intergrated intensity as a function of $\cos^2\theta$, and the waveguide geometry used in this experiment.

ments were performed using the pump and probe technique, in which the chopped ($\nu = 1700$ Hz) pump laser light impinged on the sample along the [001] direction, as shown in the inset of Fig. 1. The probe beam from a glo-bar lamp was dispersed by a 0.25-m monochromator with a resolution of ~ 3 meV in the pertinent range of 0.1–0.23 eV. After traversing the sample it was detected by a HgCdTe detector. Both the transmission, T, and the photomodulated transmission, ΔT, were recorded using a phase-sensitive, lock-in technique. The system response was accounted for by taking the ratio $(-\Delta T/T)$ which equals the change in the sample absorbance. In these experiments the sample was mounted on an evacuated cold-finger assembly of a variable-temperature cryostat. For the visible PL measurements at 2 K we have utilized an immersion-type cryostat and monitored the signals with a double monochromator having a 0.1-meV resolution.

Figure 1 shows a series of PA spectra taken at a cold-finger temperature of 90 K with the electric field of the ir radiation at an angle θ with respect to the z axis. Figure 1(a) shows the spectrum taken with an unpolarized ir beam. Two lines are observed: Line I at 180 meV and line E at 150 meV, both having $\simeq 6$ meV full width at half maximum. Figures 1(b)–1(d) show that line I is polarized along the growth direction while line E is not. The $\cos^2\theta$ dependence of line I (shown in the inset) is exactly that expected for an intrinsic transition between electronic levels with $\Delta n = 1$. Therefore, it unambiguously shows that line I originates from intrinsic, intersubband transitions within the QW. We have also measured the dependence of the PA strength on the laser intensity (I_L) and on temperature. Line I increases linearly with I_L, characteristic of intrinsic, monomolecular-type recombination of the absorbing entity. Line E, on the other hand, increases approximately as $I_L^{0.5}$ and saturates for $I_L \gtrsim 0.1$ W/cm^2, a behavior which is characteristic of extrinsic, multimolecular-type recombination.[8] The two absorption bands differ also in their temperature dependence: Whereas line E monotonically decreases as the temperature increases, line I increases slowly with increasing temperature up to $\simeq 90$ K (factor of $\simeq 3$ change in the range 20–90 K), and then decreases slowly (again by a factor of $\simeq 3$) as the temperature is increased to 150 K. In a preliminary report,[9] we have argued that the polarization, transition energies, and the temperature dependence of line E suggest that it originates from an interface-bound exciton. In the following we will prove that line I is the absorption from the exciton level ($e1$:hh1) to the exciton level ($e2$:hh1) of the photogenerated electron–heavy-hole excitons.

In order to obtain the electronic subband structure of the MQW, we have measured the photoluminescence and its excitation spectrum. Figure 2(a) shows the PL spectrum taken at 2 K and under selective excitation. The strongest PL line appears at 1.629 eV, and is

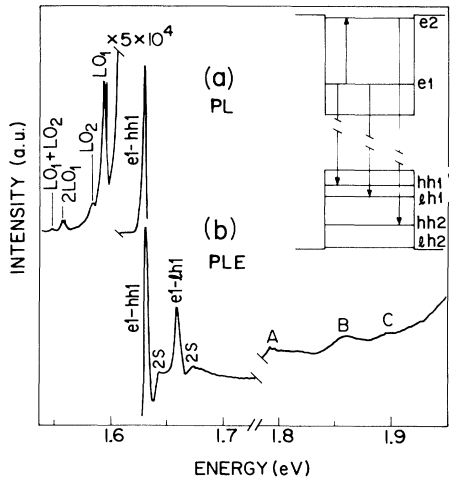

FIG. 2. (a) The PL spectrum of GaAs/$Al_{0.33}Ga_{0.67}As$ MQW and (b) the PLE profile, taken at 2 K. Inset: The schematic energy-level diagram for electrons (e), light holes (lh), and heavy holes (hh). The $2S$ exciton transitions associated with $e1$-hh1 and $e1$-lh1 lines are marked. The LO-phonon sidebands associated with the PL are also shown.

identified as the $e1$-hh1 exciton transition as shown in the schematic energy-level diagram (inset, Fig. 2). This line has a weak, low-energy shoulder which corresponds to a recombination of excitons bound to impurities. It also features several very-low-intensity LO-phonon sidebands. They are indicated as LO_1 for GaAs-like phonons and LO_2 for AlAs-like phonons (the origin of these sidebands will be discussed separately). Note that the total emission intensity below 1.62 eV is less than 10^{-3} of the 1.629-eV line. The excitation profile of the $e1_\$(hy$hh1 line is shown in Fig. 2(b). The two strongest lines are identified as the $e1$-hh1 ($1S$) and the $e1$-lh1 ($1S$) exciton transitions, respectively. Their corresponding $2S$ levels are indicated in the figure.[10]

In order to establish the connection between the infrared PA and PLE spectra, we have measured the PA excitation profile. Figures 3(a) and 3(b) show the photoexcitation spectra of line I and the $e1$-hh1 PL line, both taken at 90 K. It is clear that the PA follows exactly the photogeneration of the free excitons, indicating again that it is due to photodegenerate intersubband exciton transitions. Furthermore, we have used Bastard's[11] envelope-function approximation in order to calculate the electron and hole subband energies. Miller, Kleinman, and Gossard[12] have done this for quantum wells with widths down to about 50 Å. They found that by using a single set of QW parameters (i.e., effective masses and band offset ratio), they could obtain a good fit to the observed transitions for well widths $d > 70$ Å, but deviations occurred for narrower widths. We adopted their parameters as starting values and used the line-I energy to find the conduction-band offset, V_e (for fixed m_e and

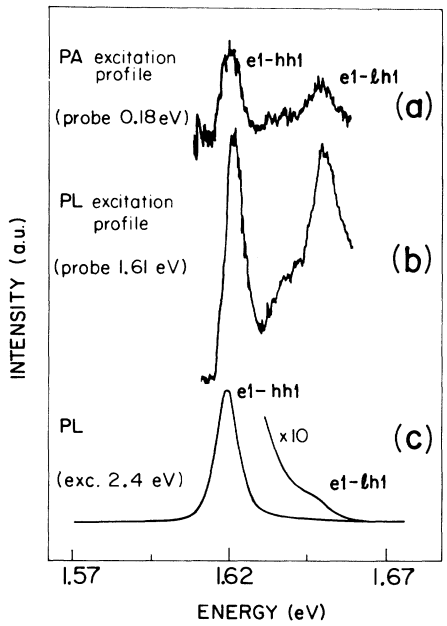

FIG. 3. The excitation profiles (a) of line I and (b) of the $e1$-hh1 PL line, taken at 90 K. The PL spectrum taken at 90 K, in the same cryostat and under the same conditions, is shown in (c).

d). Then for fixed m_{lh} we used the $e1$-lh1 exciton transition to obtain V_h, the valence-band offset. We then fitted the heavy-hole mass to obtain the energy of the $e1$-hh1 transition. Repeating this procedure, we obtained the best fit with the following QW parameters: $m_e=0.067m$, $m_{hh}=0.20m$, $m_{lh}=0.087m$, $d=49$ Å, total offset of $V\equiv V_e+V_h=475$ meV, and offset ratio of $Q=V_e/V=0.56$. The mass values, especially that of the heavy hole, and Q are somewhat different from those used by Miller, Kleinman, and Gossard,[12] but yield excellent agreement with the $e1$-hh1 and $e1$-lh1 transitions in our 50-Å wells (see Table I). Since the transitions which are observed in the PLE spectrum [Fig. 2(b)] involve excitons, while the calculated ones refer to bare e-h transitions, the binding energy of the exciton (E_{BX}) has to be considered. For the ($e1$:hh1) and ($e1$:lh1) excitons $E_{BX}\simeq 10$ meV.[10,13] However, for the ($e2$:hh1) exciton, it was neither measured nor calculated. For (en:hh$n$) ($n>1$) excitons, values of E_{BX} were estimated to be in the range 3–8 meV.[14,15] We have performed the fitting procedure assuming the extreme cases of $E_{BX}=3$ and 10 meV for the ($e2$:hh1) exciton. Table I gives the measured and calculated transition energies taking $E_{BX}=10$ meV for both excitons. Then, using the transition energies of Table I, we can identify the transitions A, B, and C of Fig. 2(b) as those between confined levels and the cladding (cl) quasi-continuum levels. If we take $E_{BX}=3$ meV for the ($e2$:hh1) exciton, we obtain negligible changes in the QW parameters while the A, B, and C (width $\simeq 20$ meV) transition energies change by less than 15 meV. The difference in gap energies of $V=475$ meV obtained here corresponds to a barrier composition of $x=0.34$ if we assume the empirical formula of Aspnes *et al.*,[16] or $x=0.29$ if we use the analysis of Monemar, Shin, and Pettit[17] of their reflectance measurements at 4 K. In any case, the two values of x are close to the nominal value of $x=0.33$.

We now consider the total integrated absorbance (I_A) of the ir transition between the $e1$ and $e2$ electron levels. For light polarized with the electric field along z, it is given by[4,18]

$$I_A=e^2hfN/4\epsilon_0 m^* cn\,, \tag{1}$$

where n is the refractive index, m^* is the effective mass, and N is the total areal carrier density. For the intersubband absorption in n-doped MQW's,[4] the oscillator strength $f\simeq 1$. Note that in these experiments there are no holes, and therefore the transitions are purely electronic. For line I, observed at an internal incident angle of 45° with respect to z, with unpolarized light, $I_A\simeq 2\times 10^{-6}$ eV at $\simeq 90$ K and 1.4×10^{-6} eV at $\simeq 30$ K. This I_A is obtained for a pump beam with a photon flux of $g\simeq 2\times 10^{17}$ cm^{-2}sec^{-1}. For a unity quantum efficiency of exciton generation, the steady-state exciton density is given by $N_{ex}=Ag\tau$ (τ is the exciton lifetime and $A\simeq 0.55$ corrects for reflections from the window and sample surfaces). Assuming, first, that line I is due to free electrons formed by exciton dissociation, we can use Eq. (1) to obtain experimentally the oscillator strength f or electronic lifetime associated with the

TABLE I. The experimental transition energies (Figs. 1 and 2), compared with those obtained from the MQW subband calculations. All interband transitions were corrected for exciton binding energy of 10 meV. The assignments of the A, B, and C transitions are given in footnotes a, b, and c, respectively; cl means transition to or from the unconfined cladding states.

	$e2$-$e1$ (eV)	$e1$-hh1 (eV)	$e1$-lh1 (eV)	A (eV)	B (eV)	C (eV)
Expt.	0.180	1.631	1.659	1.79	1.86	1.90
Calc.	0.180	1.631	1.659	1.80[a]	1.84[b]	1.92[c]

[a] $e1$-cl and $e1$-lh2.
[b] cl-lh1 and $e2$-lh1.
[c] $e2$-hh2 and cl-hh2.

$e1$-$e2$ transition. Since the electron density $N_e < N_{ex}$, Eq. (1) (corrected for the ir light geometry and polarization) yields an experimental lower bound for the quantity $f\tau$: $f\tau > 1.2\times10^{-7}$ sec at $\simeq$90 K and $> 8\times10^{-8}$ sec at $\simeq$30 K. Using $f\simeq1$ for the electronic $e1$-$e2$ transition,[4] we obtain $\tau > 10^{-7}$ sec for the free electrons. At present, there is no experimental evidence for a large density of long-lived free electrons in MQW's. If, on the other hand, we assume short decay times for the free electrons, say $\tau\sim1$-5 nsec, then $f > 20$-100. This value is much larger than the value $f\simeq1$ found for n-doped MQW's.[4] Furthermore, since the exciton binding energy is $\simeq$10 meV, there should be a strong temperature dependence in the range 10-100 K, which is not observed experimentally (see above). Thus, it is unlikely that the electronic $e1$-$e2$ transition is at the source of line I.

Alternatively, we consider the origin of line I to be excitonic; i.e., it is the excitonic transition ($e1$:hh1) to ($e2$:hh1). In this case, in analogy with interband excitonic transitions,[7] Eq. (1) is modified with $N=N_{ex}$ and $f=f_{ex}$, defined now as an "effective excitonic oscillator strength." We now obtain $f_{ex}\tau\simeq1.2\times10^{-7}$ sec at $\simeq$90 K and 8×10^{-8} sec at $\simeq$30 K. The reported[7] exciton lifetime at $\simeq$30 K is $\tau\simeq2$ nsec which yields $f_{ex}\simeq40$, while at $\simeq$90 K with $\tau\simeq5$ nsec, we have $f_{ex}\simeq25$. Note, that this value of f_{ex} is critically dependent on the value we assumed for τ. In principle, it is possible that the photoinduced transition originates from a small number of long-lived excitons. This possibility is unlikely, however, since long decay tails with sufficient strength were not observed in MQW's. We thus conclude that line I originates from intrinsic, short-lived excitons which give rise to the large oscillator strength. We note that the decrease in I_A for $T > 90$ K can now be explained by partial dissociation of the excitons.

In recent theoretical calculations of the oscillator strength of interband transitions in QW structures,[5-7] it was shown that proper consideration of the exciton wave functions gives rise to an enhancement of the transition strength by a factor of A_c/A_x. Here A_x is the area of the two-dimensional exciton and A_c is the "coherence area" of the exciton before scattering. Applying this relation to the intersubband transitions, we obtain for our sample a coherence area which is more than 25 times the area covered by the Bohr radius of the exciton.

In conclusion, we have shown that the observed infrared PA in our MQW arises from excitonic transitions with enhanced oscillator strength. Based on theoretical calculations we estimated the coherence length of the exciton center-of-mass motion to be of the order of $\simeq$5 exciton radii, or $\simeq$25 Å.

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