Excitonic absorption in modulation-doped $GaAs/Al_xGa_{1-x}As$ quantum wells

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We have performed transmission measurements in a range of undoped through heavily-modulation-doped $GaAs/Al_xGa_{1-x}As$ multiple-quantum-well structures. The observed absorption spectra demonstrate quenching of the excitonic oscillations with increasing quasi-two-dimensional electron gas. The electron density corresponding to the total bleaching of the lowest excitonic oscillation is greater than or equal to 3×10^{11} cm^{-2} for a quantum well size of 200 Å. Theoretical calculations of the absorption spectra which include the effect of carrier screening have been made. The results show that both long-range and short-range many-body effects should be included to explain the experimentally observed spectra. In the modulation-doped case, we conclude that the phase-space filling and exchange of the electron gas are the dominant effects on excitonic absorption. From the observation that the linewidth increases with the electron density, we demonstrate that the exciton lifetime reduces due to the interaction between the electrons and the excitons.

I. INTRODUCTION

The absorption spectra of high-quality GaAs/$Al_xGa_{1-x}As$ multiple-quantum-well structures (MQW's) generally show sharp excitonic structures up to room temperature due to the confinement of the carriers. Recently, research on many-body interactions in MQW's has revealed interesting phenomena, such as the excitonic nonlinear-optical effects.[1] Potentially, these effects can be utilized in integrated optics and optical data processing.[2] Many-body interactions in MQW's include electrons, holes, and unbound and bound electron-hole pairs (electron-hole plasma and excitons). In the photoabsorption experiment, a strong laser pump is used to create high density of electron-hole plasma or excitons, depending on nonresonant or resonant pumping, respectively. Carriers so created screen excitonic oscillations in absorption spectra. Several mechanisms are responsible for such a phenomenon, the phase-space filling, the correlation, and the exchange interactions. It has been argued[3] that Coulomb screening of excitons by the electron-hole plasma is relatively weak in MQW's but that the consequences of phase-space filling and exchange are significant.

Modulation-doped structures have been widely used in high-speed electronic devices. They are extensively investigated in order to handle the behavior of excitonic transitions in MQW's in the presence of high-density electron (for *n*-type) or hole (for *p*-type) gas, because the ionized carriers and their host impurities are spatially separated. The charged carriers can be controlled by appropriately doping the barriers without the need for optical pumping, which also has the desirable effect of having the states below the Fermi level filled at low temperatures. Thus, the electron (or hole) gas in quantum wells is an ideal quantum liquid[4] system which can be used to investigate the many-body interactions. Since the phase-space filling affects only the subband states below the Fermi level while the Coulomb screening should have a similar effect on all subbands, it is possible to distinguish these two different mechanisms from the excitonic absorption associated with different subbands.

Detailed theoretical calculation of the absorption spectra in modulation-doped MQW's has recently been reported.[5] In this paper we report a systematic experimental investigation of transmission in modulation-doped $GaAs/Al_xGa_{1-x}As$ (from undoped to heavily *n* type) MQW's. We have estimated the electron densities necessary for bleaching the lowest subband excitonic transitions. The results are in accord with theoretical calculations reported by Sanders and Chang.[5] We found that the Coulomb correlation, as well as the phase-space filling and the exchange, must all be included to explain the experimental results.

II. EXPERIMENT

The modulation-doped $GaAs/Al_xGa_{1-x}As$ MQW structures studied here were grown by molecular-beam epitaxy (MBE) on n^+-type or semi-insulating (100) GaAs substrates. The growth sequence consisted of 0.5 μm of GaAs buffer, 500 Å AlAs, followed by MQW's. The MQW's consisted of 24 periods with a barrier width (L_b) of 150 Å and a well width of 200 Å. The AlAs mole fraction in the barrier was about 0.3. The central part (90 Å) of the barrier layers was selectively doped with Si at doping levels ranging from 3×10^{16} to 3×10^{18} cm^{-3}, corresponding to surface densities of electrons in one well of about 2.7×10^{10} to 2.7×10^{12} cm^{-2} at low temperature since almost all of the electrons in barriers transfer to wells to keep the Fermi level constant over the crystal.[6] Figure 1 shows the structure of a typical sample along with the energy-band diagram in real space. An unintentionally doped MQE with barrier width of 100 Å, well width of 210 Å, and background impurity concentrations of about 3×10^{14} cm^{-3} (acceptor) was also used for com-

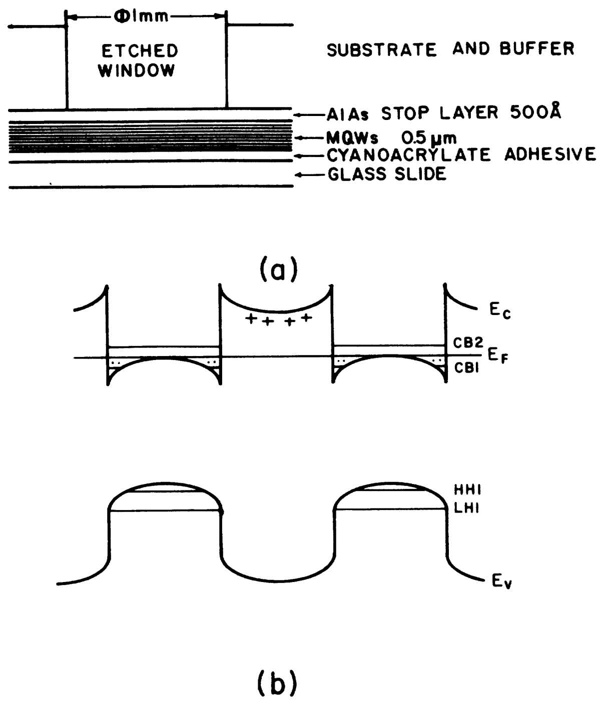

FIG. 1. (a) Modulation-doped multiple-quantum-well structure with etched window for optical transmission measurements. (b) Schematic diagram of the energy-band edge for modulation-doped structures.

parison. A very thin etch-stop layer (500 Å of AlAs) was included between the epilayers and GaAs buffer layer to facilitate the selective removal of the substrate for optical transmission measurement. All samples used in this study are listed in Table I.

Optical transmission samples were fabricated by permanently mounting the specimen onto a clear glass cover slide using a transparent cyanoacrylate adhesive. After lapping the substrate to a thickness of about 100 μm, a mechanical-chemical polish in bromine-methanol solution was used to thin the sample down to about 30 μm. Then, a circle with a diameter of 1 mm was photolithographically defined within which the entire substrate was selectively etched away. Thus, in this circular area only the MQW structure and the etch-stop remain. Upon completion, transmitted light was clearly visible.

Transmission measurements were performed with samples cooled to 3 K in a Janis optical cryostat. A broadband low-power ($\sim$20 W) tungsten lamp was used as the light source. Transmitted light was focused onto the entrance slit of 1.26-m focal length grating spectrometer; the dispersed light was detected with a cooled GaAs photocathode photomultiplier. An etched window with no MQW was also used to measure the light source spectrum so that the absolute absorption coefficient can be calculated.

The absorption spectra were obtained from transmission data using the well-known relationship

$$T=\frac{(1-R)^2e^{-\alpha d}}{1-R^2e^{-2\alpha d}} \tag{1}$$

where T is the ratio of the transmitted light through the sample (including the glass slide) and that transmitted through the clear window (glass slide with no sample). The parameter α is the absorption coefficient and d is the total absorption thickness which is 0.5 μm for our modulation-doped MQW's. The value R (reflectance) is assumed to be constant and estimated from $R=|(\sqrt{\epsilon}-1)/(\sqrt{\epsilon}+1)|^2$ to be 0.32 for GaAs ($\epsilon=13.1$). This value agrees well with the experimental data at energies much lower than the lowest absorption subband ($\alpha=0$). The reflectance at the heterointerface is neglected since the dielectric constants of $Al_xGa_{1-x}As$ and GaAs are nearly identical.

III. RESULTS AND DISCUSSIONS

Transmission spectra obtained from sample 3 with electron density of 5.4×10^{10} cm^{-2} recorded directly from the experiment are shown in Fig. 2. As can be seen the allowed excitonic transitions HHn-CBn (LHn-CBn) up to $n=5$ subbands can be clearly resolved. The notation HHn-CBm (LHn-CBm) denotes an excitonic transition with the character of the nth heavy (light) hole and mth electron subbands. The assignments were based on the theoretical calculations using the effective-mass approximation by solving the one-dimensional Schrödinger equation for a particle in an isolated finite well with proper boundary conditions and symmetries.[7] Sample parameters were determined from the MBE growth rates. In addition to the allowed transitions, the forbidden transitions like HH3-CB1 are also observed.

Using Eq. (1) we calculated the absolute absorption coefficient. Figure 3 shows the absorption spectra for all of the samples listed in Table I. The observation can be summarized as follows. (1) The absorption coefficient corresponding to excitonic transitions changes dramati-

TABLE I. Sample parameters used in this study.

Sample	L_z (Å)	L_b (Å)	X	N_D (cm^{-3})	N_s (cm^{-2})
1	210	100	0.3	undoped	
2	200	150	0.3	3.0×10^{16}	2.7×10^{10}
3	200	150	0.3	6.0×10^{16}	5.4×10^{10}
4	200	150	0.3	1.0×10^{17}	9.0×10^{10}
5	200	150	0.3	3.0×10^{17}	2.7×10^{11}
6	200	150	0.3	3.0×10^{18}	2.7×10^{12}

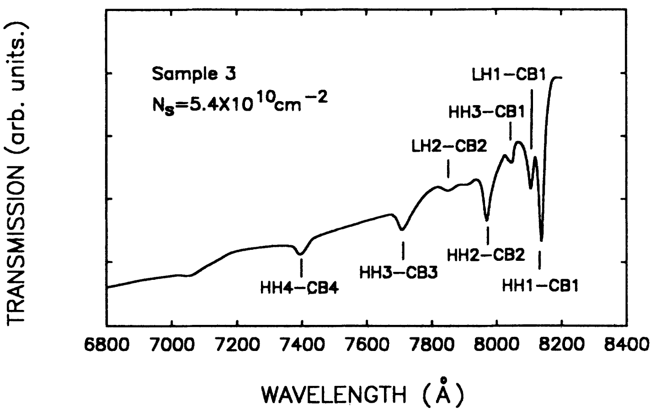

FIG. 2. Transmission spectrum of sample 3 with electron density of 5.4×10^{10} cm^{-2}.

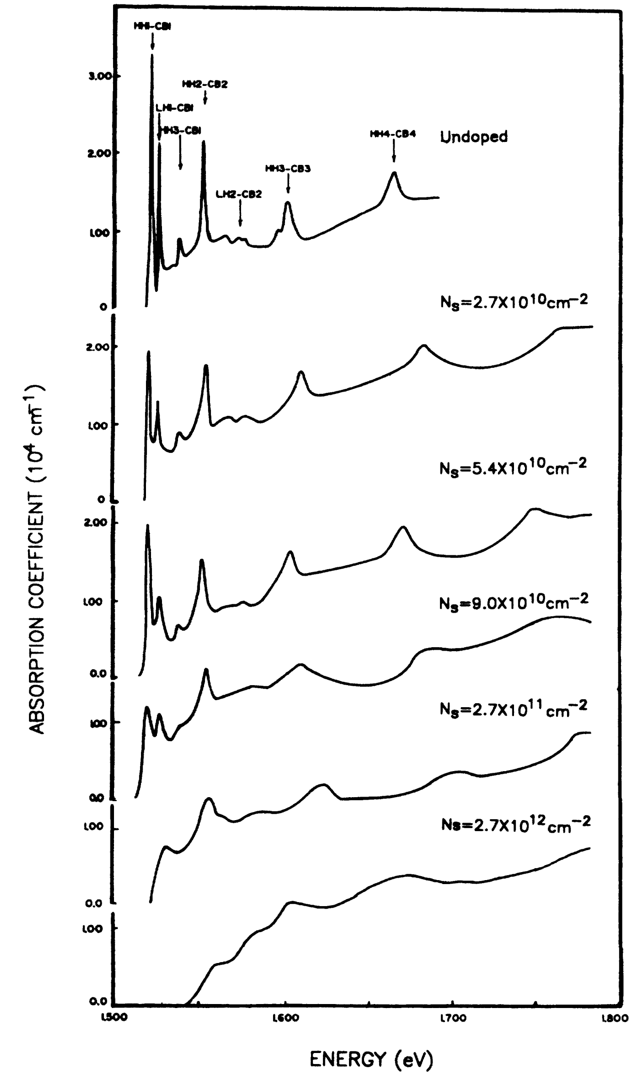

FIG. 3. Absorption spectra for all of the samples with different electron densities as listed in Table I.

cally with the electron density. When the electron density goes up to 3×10^{11} cm^{-2}, the first heavy- and light-hole associated excitonic peaks are mixed together and almost totally bleached. (2) The change in the absorption coefficient for the lowest subband excitons is more rapid than that for higher subbands with increasing electron density. (3) With the increased electron density the line shapes of the excitonic peaks become broader. (4) No consistent energy shift for $n=1$ excitons was observed. However, for the samples with high electron gas density, the excitonic transitions related to higher subbands ($n=2,3,4$, etc.) shift to higher energies.

An effective-mass method was used to calculate the absorption coefficient of excitons associated with unfilled subbands for some of the samples studied here. The calculation is similar to the one reported in Ref. 5, except here we ignore the mixing of heavy-hole and light-hole states. The effect of modulation doping is modeled by a parabolic potential of height V_0, in addition to the finite square wells, as indicated in Fig. 1. The relation between V_0 and the two-dimensional carrier concentrations is given by $N=(1/4\pi e^2)(8\epsilon \mid V_0 \mid w)$ where ϵ is the static dielectric constant of the well material, w is the well width, and e is the electronic charge of the carrier. We first solve the free-carrier problem in the envelope-function approximation for electron and holes separately. The free-electron and hole carrier states interact through the screened Coulomb force to form excitons. The screened Coulomb potential is approximated by an empirical expression given in Ref. 5. The exciton bound states and continuum wave functions are made up of linear combinations of direct products of quantum well electron and hole eigenstates of the same wave vectors. The absorption spectrum is calculated by numerically solving the exciton Hamiltonian with a **k**-space sampling. Details of this **k**-space sampling method for calculating excitonic absorption spectra were given in Ref. 8. Figure 4 shows the absorption spectra for the $n=2$ exciton calculated from the theory for a series of samples with electron densities ranging from 2.7×10^{10} to 2.7×10^{12} cm^{-2}, along with the experimental result for comparison. All

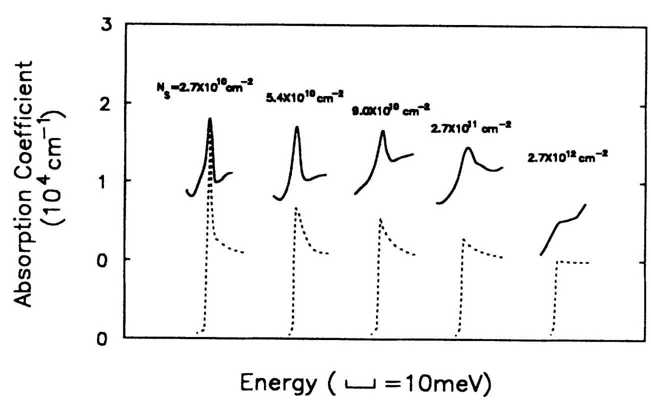

FIG. 4. Absorption spectra for $n=2$ excitonic transition for various samples. Solid line: experiment results. Dashed line: theoretical calculation.

theoretical spectra were broadened by 1.3 meV. Note that in the **k**-space sampling method, we treat the bound states and the continuum states of the exciton on equal footing. Thus there is no need to specify whether the peak structure in the absorption spectrum is due to the bound states or the final-state interaction in the continuum states (i.e., resonances). As seen in this figure, the theoretical calculation predicts a gradual decline and broadening of the excitonic peak, in qualitative agreement with the experiment. Similar results are expected for other excitons associated with the unfilled subbands. For the excitons associated with the first conduction subband which is partially filled, we expect the many-body effect to be important. Since our experiments are conducted at low temperatures (≤ 4 K), the electron gas in quantum wells is more likely to be the quantum liquid[4] and the polarization of the Fermi sea should be important. The correlation exchange, and phase-space filling effects should be considered altogether.

A more realistic calculation which takes into account the many-body effects by consideration of the single electron-hole pair excitations and the shakeup process[9] has been made and used successfully to explain the experimentally observed optical emission from electrons confined in GaAs quantum wells with polarization normal to the planes. In the calculation of the excitonic oscillations, it is found that the system has bound state for Fermi vector k_F less than 0.019 Å^{-1} if the polarization of the Fermi sea is taken into account. The relationship between the k_F and the electron density N is given by $N=(1/2\pi)k_F^2$. For $k_F=0.019$ Å^{-1}, the corresponding electron density $N=5.7\times10^{11}$ cm^{-2}, which agrees reasonably well with the experimentally observed critical density of 3×10^{11} cm^{-2}, beyond which the excitonic peak is quenched. The more realistic theoretical absorption spectra for excitonic states associated with filled subbands are being developed.

In light of the absorption spectra of Fig. 3, we note that the absorption coefficient for excitonic transitions of the lowest subband reduces more rapidly than that of higher subbands as the electron density is increased. The results are consistent with those of photoabsorption in undoped MQW's. Under high bound or unbound electron-hole pair density created by laser excitation, the similar behavior of the excitonic oscillation was observed. From the basic physical point of view, there are two mechanisms that can account for these observations. (In the photoabsorption case, these phenomena are well known as the nonlinear-optical effects.) One is the long-range Coulomb screening of excitons by the carriers, and the other is the phase-space filling and exchange. It is believed that the phase-space filling and exchange are significant, as has been demonstrated by our calculation and by other researchers.[3] Our experiments make this argument more clearly since our measurements were performed at low temperatures and only the lowest conduction subband is partially occupied by the electrons. In fact, the more rapid change of the absorption coefficient associated with the lowest subband gives strong evidence that the phase-space filling and exchange are more important than the screening effect which should affect all of the subbands. If we estimate from Fig. 3 that the electron density corresponding to the bleaching of the excitonic oscillation associated with the lowest subband is about 3×10^{11} cm^{-2}, while that associated with the higher subbands is 3×10^{12} cm^{-2}, we may conclude that the effect of the phase-space filling and exchange are about 10 times stronger than the effect of the Coulomb screening on the excitonic resonance.

The homogeneous broadening of the excitonic absorption peak reflects the lifetime of the excitonic oscillations. The observed full width at half maximum (FWHM) is 1.5 meV for the undoped sample (the electron density due to background impurity is estimated to be below 10^9 cm^{-2}), and rises to about 5.6 meV for the sample with electron density of about 10^{11} cm^{-2}. For the undoped MQW's, the linewidth of the absorption spectrum at low temperature is believed to be dominated by the inhomogeneous broadening resulting from thickness fluctuations. For the modulation-doped samples, however, the increase of the linewidth should be explained by the decrease of the exciton lifetime (or ionization time) due to the existence of the electron gas which leads to a smaller exciton binding energy. Figure 5 gives the experimental linewidths of the excitonic transitions associated with the first conduction and valence subbands as a function of the electron densities N. We estimated the exciton lifetime is about 0.18 ps, by using the uncertainty principle $\tau\sim\hbar/W_{\mathrm{FWHM}}$, for electron density of about 10^{11} cm^{-2}.

Theory[10] and experiment[11] have shown that the quasi-two-dimensional electrons confined in a semiconductor should shrink the band gap, known as the "gap renormalization." In our experiments, we did not observe the consistent change of the lowest excitonic transition energies with the electron densities. Two different effects contribute to the change of the excitonic resonant energy. Renormalization reduced the energy gap by several meV for electron densities smaller than 10^{11} cm^{-2}.[9] However, the screening of the electrons reduces the exciton binding energy by about the same order.[3] These two opposite effects make the net change of the exciton resonant energy very small. In addition, fluctuations in the quantum well thickness lead to the energy changes. So it is not

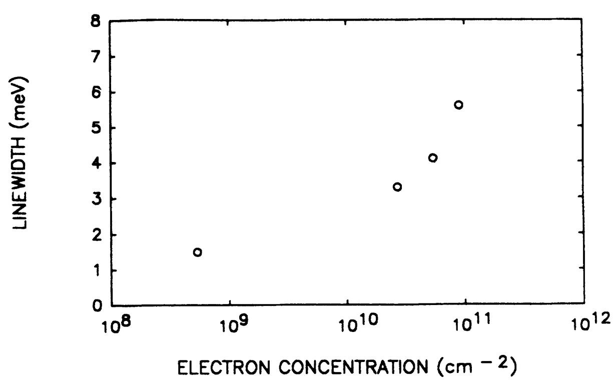

FIG. 5. Experimental linewidths of the first excitonic absorption vs electron density.

easy to compare the peak energies from different samples with different electron densities.

IV. CONCLUSIONS

We have performed transmission measurements in several $GaAs/Al_xGa_{1-x}As$ MQW's, undoped through heavily modulation doped. The observed absorption spectra demonstrate the quenching effect of the excitonic oscillations caused by the quasi-two-dimensional electron gas. The electron density corresponding to complete bleaching of the lowest excitonic oscillations is 3×10^{11} cm^{-2} for the quantum wells of 200 Å thickness. The theoretical calculations which only include the long-range Coulomb correlation and which include both correlation and exchange have been made. The results show that both long-range and short-range many-body effects should be included to explain the experimentally observed spectra. In the modulation-doped case, we conclude that the phase-space filling and exchange of the electron gas are the dominant effects on the excitonic absorption. From the observation that the linewidth increases with the electron density, we demonstrate that the exciton lifetime (or the exciton binding energy) reduces due to the interaction between the electrons and the excitons.

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