Carrier effects on the excitonic absorption in GaAs quantum-well structures: Phase-space filling

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The carrier effects on the excitonic absorption in GaAs quantum-well structures have been investigated both theoretically and experimentally. A two-dimensional model was used to calculate the oscillator strength and binding energy of excitons associated with filled subbands, with phase-space filling being taken into account. The calculation gives explicitly the oscillator strength of excitons as a function of two-dimensional carrier density. The results are compared with measured absorption data from a series of *p*-type modulation-doped GaAs/$Al_xGa_{1-x}As$ multiple-quantum-well structures, and quantitative agreement is obtained. The calculation shows that the effect of phase-space filling on the binding energy of a bound state can be described by an effective dielectric constant as a function of carrier density. It predicts the decrease of exciton binding energy with carrier density due to phase-space filling, which has been experimentally observed.

I. INTRODUCTION

It is well known that the optical-absorption spectrum of an undoped GaAs/$Al_xGa_{1-x}As$ multiple-quantum-well structure (MQWS) is dominated by sharp excitonic features at low temperatures. Recent investigations revealed that those excitonic features can be dramatically affected by free carriers appearing in quantum wells, such as electrons, holes, electron-hole plasmas, and the other excitons.[1-6] Depending on the density of carriers, the excitonic absorption can be either depressed or fully quenched. The physics behind the observed phenomena is essentially due to the many-body effects, which can be divided into three categories.[7] The first one comes from the Pauli exclusion principle. Electrons and holes which are made up of excitons are fermions. If a state in conduction band is occupied by an electron, the transition of another electron from valence band into such a filled state is blocked. This phenomenon is known as the phase-space filling.[7] The second process is the carrier screening to electron-hole Coulomb attraction.[8,9] The screening reduces the Coulomb interaction of electron-hole pair, which in turn reduces the exciton binding energy and lifetime. The last mechanism is the short-range exchange and correlation interactions among carriers.[10-13] There is some evidence that the effect of exchange and correlation interactions on the excitons in two-dimensional structures is very important, in contrast to that in three-dimensional systems.[12,13]

Among various approaches of introducing carriers into quantum wells is the modulation-doping technique.[14] The MQWS used in our study were selectively doped in the central part of barriers by Si (for *n*-type) or Be (for *p*-type). The ionized electrons (or holes) transfer from the doped part of $Al_xGa_{1-x}As$ barriers to GaAs wells to keep the Fermi level constant over the entire crystal at the thermal equilibrium. The carriers fill the lowest conduction (or highest valence) subband, and get spatially separated from their host impurities. Investigation of the modulation-doped structures shows qualitatively that the dominant mechanism responsible for quenching excitons associated with filled subbands is phase-space filling.[5,7] While for the excitons associated with empty band, the importance of screening, and exchange and correlation interactions are comparable.[7,12,13] These observations make the phase-space filling distinguishable from the other mechanisms through the study of the excitons associated with different subbands.

Although many experimental investigations have been carried out, a quantitative understanding of the many-body effects in quantum-well structures is still lacking. For example, the theoretical work on phase-space filling and exchange and correlation effects in quantum-well structures are necessary for the analysis of experimental observations. It is the purpose of this paper to investigate theoretically and experimentally the effect of phase-space filling on the excitonic absorption in GaAs/$Al_xGa_{1-x}As$ MQWS. First a two-dimensional model will be used to calculate the oscillator strength and binding energy of excitons as a function of carrier density at low temperatures. Then the experimental results from a series of *p*-type and *n*-type modulation-doped MQWS will be described. Finally, these results will be compared along with the underlying fundamentals.

II. EXCITON OSCILLATOR STRENGTH

The exciton discussed here is an electron-hole pair bonded through the Coulomb potential. It can be created in a semiconductor by photon absorption. Since the photon wave vector is very small compared to the dimension of the Brillouin zone of the semiconductor crystal, the photocreated electron in the conduction band and the hole in the valence band have essentially the same magni-

tude of momentum; in other words, the transition is vertical in $\mathbf{k}$ space. In this case, the exciton states can be written as a linear combination of the independent electron and hole states[15]

$$|n\rangle=\sum_{\mathbf{k}}\chi_{\mathbf{k}}^{n}(1-f_k)^{1/2}|\mathbf{k},c\rangle\,|-\mathbf{k},v\rangle\ , \tag{2.1}$$

where n is the quantum number denoting the different exciton states, $|\mathbf{k},c\rangle$ and $|-\mathbf{k},v\rangle$ are the free single-electron and -hole states (or Bloch states) with momentum $\mathbf{k}$ and $-\mathbf{k}$ in conduction and valence bands, respectively, $\chi_{\mathbf{k}}^{n}$ is the exciton envelope function in momentum space, and f_k is the Fermi function which is 1 for an occupied state and 0 otherwise. The factor $(1-f_k)^{1/2}$ is introduced to account for the effect of other carriers.[16] It simply reflects the exclusion principle that the excitations to the occupied states are blocked. The exciton oscillator strength (i.e., f factor) can be calculated from the probability of transition from the ground state $|0\rangle$ to the excited states $|n\rangle$,[15] and the result is given by

$$f_n=A\,|x_{cv}|^2\left|\int_k\frac{dk}{(2\pi)^2}\chi_{\mathbf{k}}^{n}(1-f_k)^{1/2}\right|^2\ , \tag{2.2}$$

where A is a constant, and x_{cv} is the dipolar matrix element between conduction and valence band.

To calculate f_n from (2.2), we need to know the exact wave function $\chi_{\mathbf{k}}^{n}$, which must be solved from the Schrödinger equation for quantum-well structures considering all of the many-body effects. This is not trivial and the exact solution is unavailable. Here we use the following approximation (similar to the Born approximation) to calculate the f factor for $1s$ excitons:

$$\chi_{\mathbf{k}}^{1s}(N)\approx\chi_{\mathbf{k}}^{1s}(N=0)\ , \tag{2.3}$$

where $\chi_{\mathbf{k}}^{1s}(N=0)$ is the wave function for an isolated exciton in an ideal two-dimensional structure, which can be solved exactly from Schrödinger equation, as

$$\chi^{1s}(r)=\left[\frac{2}{\pi}\right]^{1/2}\frac{a_0}{2}\exp\left[-2\frac{r}{a_0}\right]\ , \tag{2.4a}$$

$$\chi_{\mathbf{k}}^{1s}(N=0)=\frac{\sqrt{2\pi}a_0}{[1+(ka_0/2)^2]^{3/2}}\ , \tag{2.4b}$$

where $a_0=\epsilon\hbar^2/e^2\mu$ is the effective bulk Bohr radius with μ and ϵ being the reduced effective mass of the electron-hole pair and the dielectric constant, respectively. Substituting (2.4b) into (2.2) gives f factor as a function of N. At low temperature, we obtain

$$\frac{f_{1s}(N)}{f_{1s}(0)}=\frac{1}{1+N/N_c} \tag{2.5}$$

and

$$N_c=\frac{2}{\pi a_0^2}\ , \tag{2.5a}$$

where the relation $k_F^2=2\pi N$ has been used.

Figure 1 shows the f factor as a function of N for the first heavy-hole (1C-1H) excitons in GaAs quantum wells. The following parameters have been used: $m_e^*=0.067m_0$, $m_{\rm hh}^*=0.14m_0$, $\epsilon=13.1$. The heavy-hole effective mass $m_{\rm hh}^*$ is taken from the band-structure calculation including band mixing for GaAs quantum wells with a well width of 100 Å.[17] The results show that for carrier density $N<10^{10}$ cm^{-2}, the phase-space filling has a negligible effect on the exciton oscillator strength. For $N>10^{10}$ cm^{-2}, f_{1s} drops off rapidly with N. The N_c defined in (2.5a) is called the critical carrier density, at which

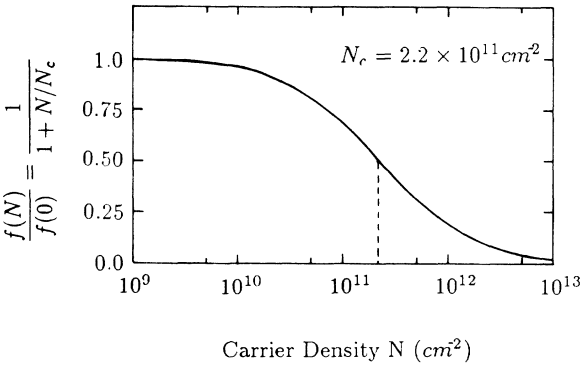

FIG. 1. Oscillator strength of 1C-1H exciton as a function of two-dimensional carrier density, calculated from a two-dimensional model considering phase-space filling.

$$f_{1s}(N=N_c)=\tfrac{1}{2}f_{1s}(N=0)\ . \tag{2.6}$$

The N_c is calculated to be 2.2×10^{11} cm^{-2} for this case.

It is interesting to note that Eq. (2.5) is equivalent for both n- and p-type structures. In other words, the critical density defined in (2.5a) is the same for both electrons and holes in our approximation. Excitons can be quenched by either electrons or holes with the same density. The critical density N_c depends only on the reduced exciton effective Bohr radius, a_0, which in turn depends on the reduced exciton effective mass μ. The result is not surprising since the photon excited electrons in the conduction band and holes remaining in the valence band have the same magnitude of wave vector (vertical transition). The number of states below this wave vector, which is close to the Fermi wave vector, is the same for both conduction and valence bands.

III. EXCITON BINDING ENERGY

The effect of phase-space filling on exciton binding energy is calculated from the Schrödinger equation, using the effective-mass approximation, in $\mathbf{k}$ space:[18]

$$(\varepsilon_{\mathbf{k}}-E)\chi_{\mathbf{k}}+\int_{\mathbf{k}'}\frac{d\mathbf{k}'}{(2\pi)^2}V_{\mathbf{k}\mathbf{k}'}\chi_{\mathbf{k}'}=0\ , \tag{3.1}$$

where $\varepsilon_{\mathbf{k}}=\hbar^2\mathbf{k}^2/2\mu$, and $V_{\mathbf{k}\mathbf{k}'}$ is the Coulomb interaction between states $\mathbf{k}$ and $\mathbf{k}'$. At low temperature, the integral over $\mathbf{k}'$ is limited to those states outside the Fermi disk for a two-dimensional structure due to the exclusion principle.[18]

If we divide the second term in (3.1) into the integral over all $\mathbf{k}'$ minus the integral over $\mathbf{k}'$ inside the Fermi disk, and take a Fourier transform, we get the wave equation in real space:

$$\left[-\frac{\hbar}{2\mu}\nabla^2-E\right]\chi(r)+(1-\xi)V_0(r)\chi(r)=0 , \tag{3.2}$$

where $V_0=-e^2/\epsilon r$ is the bare Coulomb potential and ξ is a correction to V_0 due to phase-space filling,

$$\xi(r)\chi(r)=\int_0^{k_F} dk\ \frac{k}{2\pi}\chi_{\mathbf{k}}J_0(kr) , \tag{3.3}$$

where $J_n(kr)$ is the nth Bessel function.

The exact $\chi_{\mathbf{k}}$ cannot be solved from Eq. (3.1). To analytically evaluate ξ from Eq. (3.3), we make the approximation that the perturbation on the excitons by free carriers is so small that the bound state is still reserved and the wave function of excitons still retains a similar form to Eq. (2.4), except that the Bohr radius a_0 is different. As being reported, there is always a bound state for arbitrarily weak attractive potential for a 2D structure, if only the long-range screening is considered.[19] However, this is not true if the exclusion principle is included as in our case.[20] Thus the approximation is compatible to the fact that the binding energy of a bound state is to be calculated. Mathematically, the approximation is written as

$$\frac{a_0k_F}{2}\ll 1 . \tag{3.4}$$

Using (2.4) and (3.4), Eq. (3.3) is evaluated as

$$\xi(r)\chi(r)\approx\frac{a_0}{\sqrt{2\pi}}\frac{k_F}{r}J_1(k_Fr) . \tag{3.5}$$

For small r,

$$\xi\approx\frac{1}{2}\left[\frac{a_0k_F}{2}\right]^2\ll 1 . \tag{3.6}$$

Combining (3.2) and (3.6), we finally obtain that the phase-space filling can be described by a carrier density-dependent effective dielectric constant

$$\epsilon(N)=\frac{\epsilon(0)}{1-N/(2N_c)} , \tag{3.7}$$

where N_c is the critical carrier density as defined in (2.5a). Using the effective dielectric constant, $\epsilon(N)$, the Coulomb potential, the Schrödinger equation, and the wave function have basically the same form as Eq. (2.4), but with a new a_0 associated with the density-dependent dielectric constant.

We have calculated the exciton oscillator strength again using the effective dielectric constant (3.7). The result is exactly the same as that obtained in the last section, Eq. (2.5), under the approximation of (3.4). Using (3.7), we can also get the exciton binding energy

$$E_{\mathrm{XB}}(N)=E_{\mathrm{XB}}(0)\left[1-\frac{N}{2N_c}\right]^2 . \tag{3.8}$$

This equation explicitly shows the decline of the exciton binding energy with the increase of carriers. When N becomes close to N_c, E_{XB} approaches $\frac{1}{4}$ of the value of an isolated exciton.

IV. EXPERIMENTAL RESULTS AND DISCUSSIONS

The absorption spectra for n-type modulation-doped MQWS with well width of 200 Å and various electron densities have been reported previously.[5] Here we present experimental data taken from a series of p-type modulation-doped MQWS. The samples were grown by molecular-beam epitaxy. The structural parameters of these MQWS are listed in Table I, where N_s is the hole density per well obtained from Hall measurements. The quantum-well and barrier thickness are both 100 Å, with 50 Å in the central part of barriers being doped with Be. The reason for having a narrow quantum-well thickness (100 Å) is to simulate a two-dimensional system that can be reasonably addressed by our calculation. In addition, the use of p-type samples makes it possible to compare the effect of holes in p-type samples with that of electrons in n-type samples. For this reason, we also grew an n-type sample with the same well thickness of 100 Å as p-type samples and an electron density close to the critical density. The structure parameters of this sample are also listed in Table I. The details of experiments have been described elsewhere.[5]

Figure 2 gives the absorption spectra of p-type modulation-doped GaAs/Al_xGa_{1-x}As MQWS listed in Table I. As observed, the overall picture is very similar to that from n-type samples described in Ref. 5: The absorption by excitons decreases with increasing two-dimensional hole-gas (2D HG) density. The resonant peaks also broaden with carrier density. However, there is an important difference: In contrast to the absorption

TABLE I. Modulation-doped GaAs/Al_xGa_{1-x}As multiple-quantum-well structures used in this study.

Sample No.	L_z (Å)	L_b (Å)	x	Carrier	N_s (cm^{-2})
1	100	100	0.3	hole	1.2×10^{9}
2	100	100	0.3	hole	1.4×10^{10}
3	100	100	0.3	hole	1.5×10^{11}
4	100	100	0.3	hole	7.5×10^{11}
5	100	100	0.3	electron	2.6×10^{11}

spectra from n-type samples, the first heavy- and light-hole (1C-1H and 1C-1L) excitons show the different dependence on the hole density in p-type samples. The excitons of heavy holes quench more rapidly than those of light holes, as shown in curves 3 and 4. In particular, for the sample with a 2D HG density of 7.5×10^{11} cm^{-2}, the absorption of the heavy-hole exciton is no longer observable, while the light-hole exciton is still distinguishable.

We note from the self-consistent calculation that only the 1H subband is filled with holes even for a 2D HG density of 7.5×10^{11} cm^{-2}. Our observation, therefore, supports the argument that the phase-space filling is the dominant mechanism. To demonstrate that this is really the case, we also carried out the measurements at different temperatures for sample No. 4. Since the phase-space filling depends more on temperature, through Fermi function, than screening and other many-body effects, we expect to see more dramatic change in the 1C-1H exciton than in the 1C-1L exciton with temperature. The results are shown in Fig. 3. At low temperature (4 K), there is only one absorption peak which is dominated by the light-hole exciton absorption. The heavy-hole exciton is not observable. When the temperature rises, as expected, the 1C-1H exciton peak with lower energy emerges and its intensity increases more rapidly with temperature than that of light hole. Near 130 K, the absorption due to heavy- and light-hole excitons becomes comparable.

Since the carrier effect on 1C-1H excitons is dominated by phase-space filling, screening and other mechanisms are negligible, and we can use the theoretical model developed in Sec. II to explain the dependence of 1C-1H excitonic absorption on the carrier density. To give a quantitative comparison between theory and experiments, we use a Lorentzian line shape to model the absorption spectra,

$$L(\hbar\omega)=\frac{f_{1s}(N)}{f_{1s}(0)}\frac{\Gamma/2\pi}{(\hbar\omega-E_{1s})^2+(\Gamma/2)^2}\ . \tag{4.1}$$

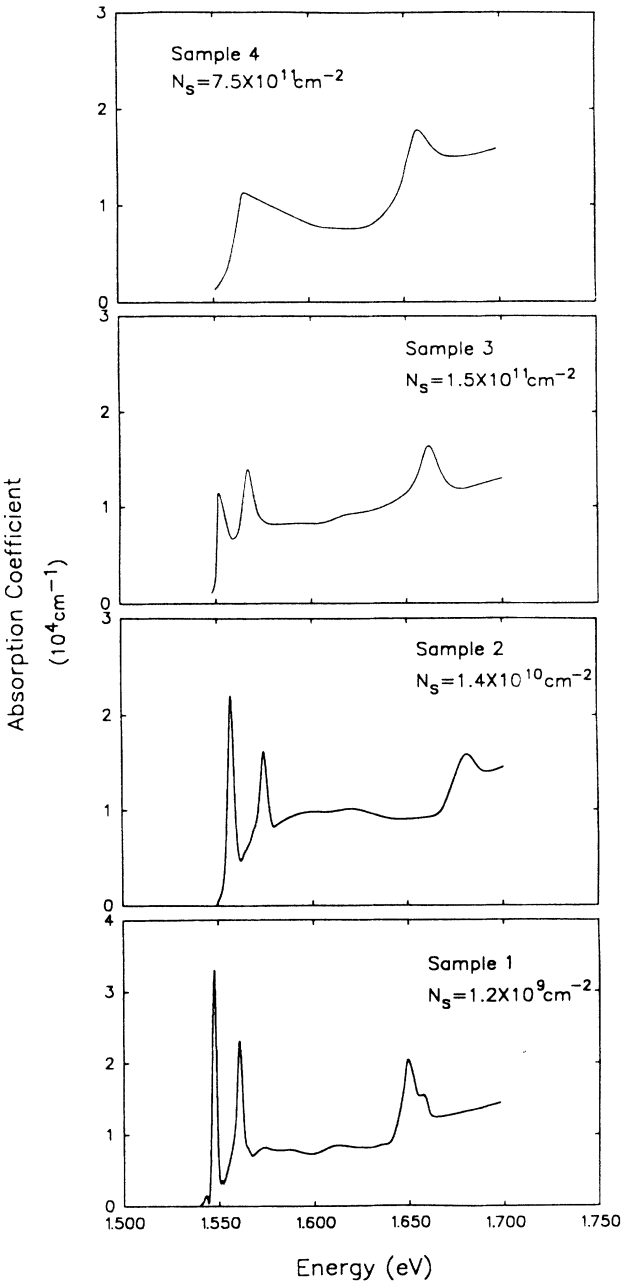

FIG. 2. Absorption spectra measured from a series of p-type modulation-doped GaAs/Al_xGa_{1-x}As multiple-quantum-well structures with different hole densities.

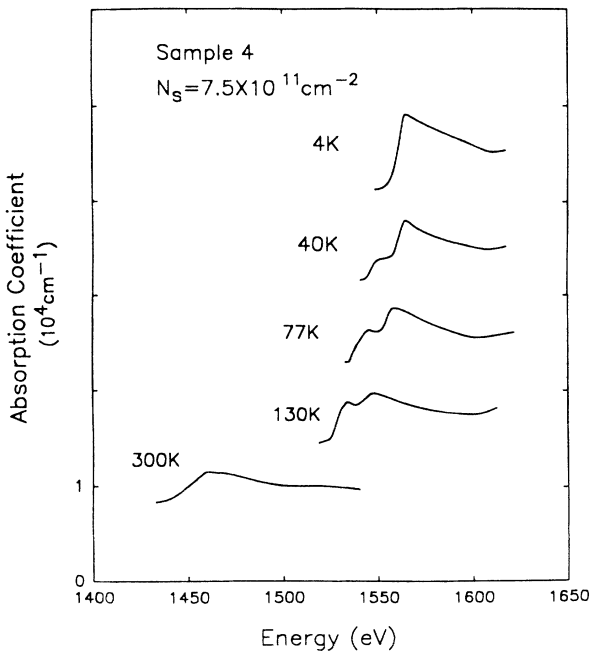

FIG. 3. Absorption spectra of a p-type GaAs/Al_xGa_{1-x}As modulation-doped multiple-quantum-well structure, measured at different temperatures.

TABLE II. Oscillator strength and resonant line broadening of the 1C-1H exciton for a series of p-type modulation-doped $GaAs/Al_xGa_{1-x}As$ multiple-quantum-well structures.

Sample No.	2D HG Density (cm^{-2})	$f(N)/f(0)$	Γ (FWHM) (meV)
1	1.2×10^{9}	0.99	2.3
2	1.4×10^{10}	0.94	4.0
3	1.4×10^{11}	0.59	5.8
4	7.5×10^{11}	0.22	8.0

The integration of Eq. (4.1) over $\hbar\omega$ gives the normalized oscillator strength $f(N)/f(0)$. The broadening parameter Γ is taken from experimental data as the full width at half maximum (FWHM), for sample Nos. 1, 2, and 3. For sample No. 4 the data are extrapolated from those of the other samples.

Table II lists the hole densities, broadening parameters, and the calculated exciton oscillator strength for a number of p-type modulation-doped samples with a well width of 100 Å. The line shapes calculated from (4.1) are shown in Fig. 4, along with the experimental absorption data for comparison. Although the theoretical model is relatively simple and there may be some uncertainty in obtaining absolute experimental data, the excellent agreement between theory and experiment lends support for the theoretical model presented.

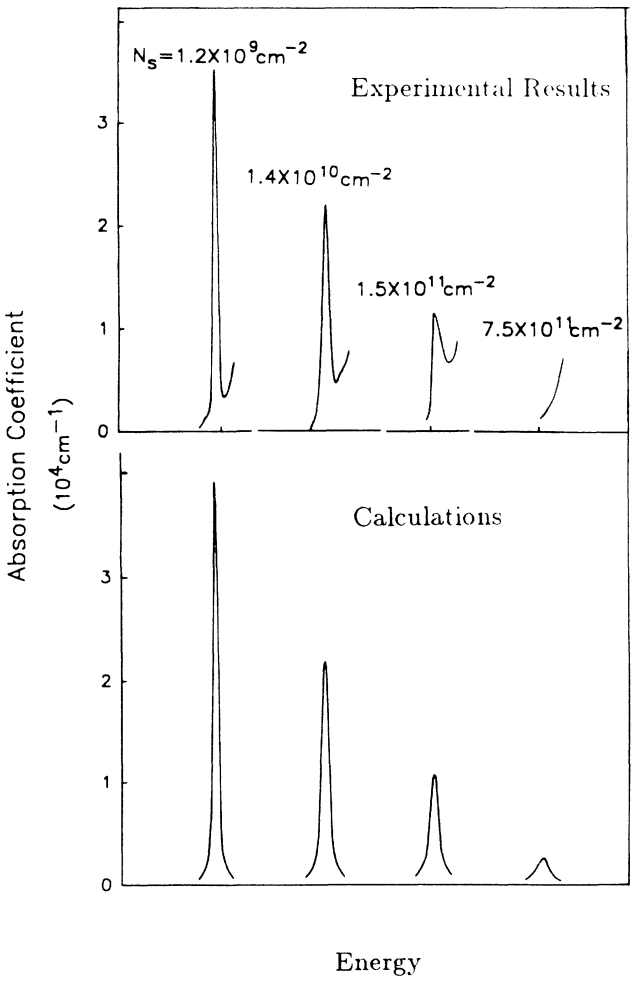

FIG. 4. (a) Measured and (b) calculated absorption coefficients for 1C-1H excitons for a series of p-type modulation-doped $GaAs/Al_xGa_{1-x}As$ multiple-quantum-well structures with different hole densities listed in Table I.

It is not easy to measure directly the change in exciton binding energy with carriers in MQWS. That measured in experiments is the peak energy for each excitonic transition associated with the different subbands in MQWS. The energies of those peaks are affected by many factors: the confinement energies, which depend mainly on the quantum-well thickness, renormalization of the band gap,[21] and exciton binding energies. Since the peak shift due to the fluctuation of quantum-well thickness and the band-gap renormalization are on the same order of magnitude as the exciton binding energy, it is difficult to obtain the binding energy accurately either by comparing the results from different samples or using the sample by photopumping to create different carrier densities. Even if the change in exciton binding energy has been subtracted from the total shift of exciton peak, it is still not clear if this change is due to the phase-space filling or the other many-body effects such as screening. Fortunately, there are two important properties of phase-space filling, which make it possible for us to distinguish this phenomenon from the other effects: (1) Phase-space filling only affects the excitons associated with filled subbands, but not the empty subbands; and (2) it is strongly temperature dependent. Most of the other mechanisms, such as band-gap renormalization, screening, and the well thickness fluctuation equivalently affect all of the subbands and are less sensitive to temperature.

Using the above properties, we can probe the change in exciton binding energy due to phase-space filling (PSF), at least qualitatively, as follows. We first measure the peak energies of 1C-1H and 1C-1L excitons as a function of temperature. Then we take the energy difference between 1C-1H and 1C-1L exciton peaks, which is given by

$$\Delta E(T)=E_{\text{1C-1L}}-E_{\text{1C-1H}}$$
$$=\text{const}+(\Delta E^{\text{PSF}}_{\text{1C-1L}}-\Delta E^{\text{PSF}}_{\text{1C-1H}})\,, \qquad (4.2)$$

where the constant describes the temperature-insensitive parts such as the confinment energies, the exciton binding energies and their changes due to screening and other many-body effects, etc. The temperature dependence of the band gap is canceled since it is identical for both heavy- and light-hole excitons. The only temperature-dependent terms are the changes in exciton binding energies due to phase-space filling, $\Delta E^{\text{PSF}}_{\text{1C-1L}}$ and $\Delta E^{\text{PSF}}_{\text{1C-1H}}$. For n-type samples, the electrons fill the first conduction subband and the phase-space filling affects both 1C-1H and 1C-1L excitons. In this case, it is expected that $\Delta E^{\text{PSF}}_{\text{1C-1L}}$ and $\Delta E^{\text{PSF}}_{\text{1C-1H}}$ are very close and both terms are canceled. In other words, the shift in heavy-hole exciton peak with temperature must follow that of light hole. For the p-type sample, however, only the first heavy-hole subband is occupied, while the light-hole subband is empty, or $\Delta E^{\text{PSF}}_{\text{1C-1L}}=0$. In this case, we should see a temperature

dependence of ΔE, which is dominated by the changes in the binding energy of the first heavy-hole excitons due to phase-space filling, $\Delta E^{\mathrm{PSF}}_{\mathrm{1C\text{-}1H}}(T)$.

We measured the exciton peak energy as a function of temperature for two samples, Nos. 3 and 5. The structure parameter of sample No. 5 is an n-type modulation-doped MQWS with electron density per well of 2.6×10^{11} cm^{-2}, as listed in Table I. Sample No. 3 is p-type with hole density of 1.5×10^{11} cm^{-2}. Both samples have nominally identical well width of 100 Å. Figure 5 shows the relative energies of the exciton peak changing with temperature. The energies for light-hole excitons have been rigidly shifted to make the comparison easy. As we expected, the transition energy for heavy holes precisely follows that for light holes in the n-type sample. This, however, is not true for the p-type sample at low temperatures. The higher peak energy for heavy-hole excitons than that for light-hole excitons reflects the lower binding energy for the former due to the phase-space filling.

Quantitatively, the energy difference taken from Fig. 5 at $T=0$ for p-type samples is 1.3 meV, which is much smaller than $\Delta E_{\mathrm{XB}}=E_{\mathrm{XB}}(N)-E_{\mathrm{XB}}(0)$ calculated from Eq. (3.8). The main reason is that Eq. (3.8) is no longer applicable since the carrier density for sample No. 3 is close to that of N_c and the assumption (3.4) leading to (3.8) is not valid. It may also be due to the nonparabolicity of valence subbands, especially for the light hole, which was not considered in our calculation. Nevertheless, it is clearly evident from the results shown in Fig. 5 that the binding energy decreases due to phase-space filling for heavy-hole excitons.

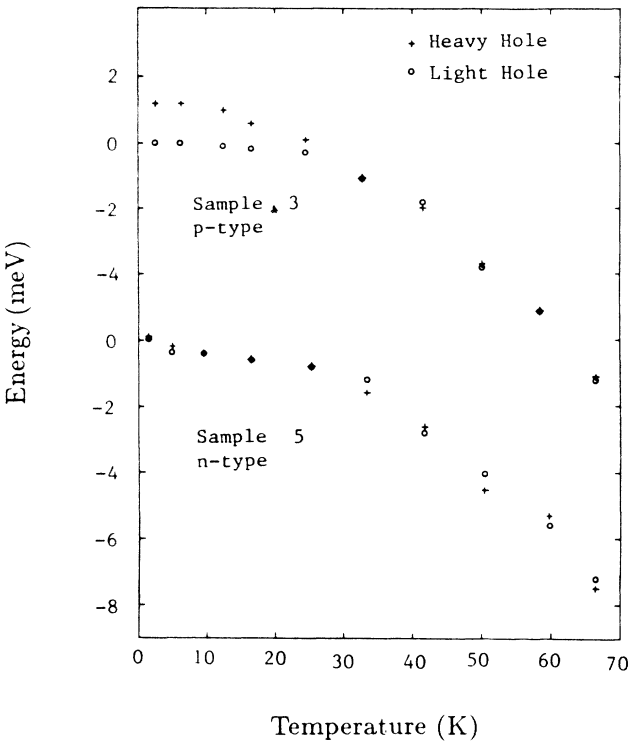

FIG. 5. Peak energies of 1C-1H and 1C-1L excitons changing with temperature for (a) n-type and (b) p-type modulation-doped $\mathrm{GaAs/Al}_x\mathrm{Ga}_{1-x}\mathrm{As}$ multiple-quantum-well structures.

V. CONCLUSIONS

The effects of free electrons and holes on the excitonic absorption in $\mathrm{GaAs/Al}_x\mathrm{Ga}_{1-x}\mathrm{As}$ modulation-doped MQWS have been investigated. The absorption spectra show that the quenching of excitons associated with the filled subbands can be observed at the carrier densities of a few 10^{10} cm^{-2}, for both electrons and holes. The dominant mechanism responsible for quenching is the phase-space filling, while the screening and other many-body effects are only important for the excitons associated with empty subbands. A two-dimensional model considering phase-space filling has been developed to calculate the oscillator strength and binding energy of excitons. The calculation gives explicitly the exciton oscillator strength as a function of two-dimensional carrier density. It shows that at a carrier density of 2.2×10^{11} cm^{-2}, the same for both electrons and holes in GaAs, the exciton oscillator strength reduces to half of that in an updoped quantum well at low temperatures. The calculated results have been compared with measured absorption data from a series of p-type modulation-doped $\mathrm{GaAs/Al}_x\mathrm{Al}_{1-x}\mathrm{As}$ MQWS, and quantitative agreement is obtained. The calculation also predicts the decrease of exciton binding energy with carriers due to phase-space filling. It is shown that the effect of carriers on the exciton binding energy due to phase-space filling can be described by an effective dielectric constant, which depends on the carrier density. The predicted change of exciton binding energy has been experimentally demonstrated.

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