Onset of exciton absorption in modulation-doped GaAs quantum wells

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We study the evolution of the absorption spectrum of a modulation-doped GaAs/$Al_xGa_{1-x}As$ semiconductor quantum well with decreasing the carrier density. We find that at some critical electron density there is a sharp change in the line shape and the transitions energies of the exciton peaks. We show that this critical density marks an abrupt transition from a simple excitonic behavior to a Fermi edge singularity.

The absorption spectrum in the presence of a Fermi sea of electrons has been a subject of theoretical and experimental research for more than three decades. The interest in this problem was triggered by the pioneering work of Mahan,[1] who showed that in metals and bulk semiconductors the onset of the absorption spectrum should exhibit a power-law singularity of the form $(\omega-\omega_0)^{-\alpha}$. This singularity, which became known as the Fermi edge singularity (FES), reflects the final-state response of the Fermi sea electrons to the attractive potential of the valence-band hole. Mahan's work was followed by other theoretical works, which established the many-body nature of the FES and provided the tools to treat it.[2,3] The FES was indeed observed in the x-ray absorption of metals and in the interband absorption of semiconductors. In semiconductor heterostructures it is manifested as a pronounced enhancement of the absorption threshold, with an asymmetrical line shape: a fast rise at the low-energy side and a slow fall at high energies.[4–6] The high-energy tail is due to photon absorption, which is mediated by shake-up processes in the electron gas.

An intriguing aspect of the behavior of a system of a two-dimensional electron gas (2DEG) and a hole is the existence of a bound state. It is well known that in two dimensions bound states occur for arbitrarily small attractive potentials. Thus, the attractive potential of a valence hole should have a bound state even after the many-body interactions have been accounted for. This bound state was a subject for several recent theoretical investigations.[7–9] It was found that it is doubly occupied by electrons with opposite spins and should persist to a high electron density. It was predicted that the bound state should be manifested in the absorption spectrum of semiconductor quantum wells containing a 2DEG as two peaks, which correspond to final states where the bound state is doubly or singly (ionized) occupied.[7,9,10] The energy difference between the two peaks should therefore be $\Delta E=E_F+E_B$, which is the cost of energy to excite an electron from the bound state to above the Fermi energy (E_F is the Fermi energy and E_B is the electron binding energy). Indeed, these two peaks were recently observed in CdTe modulation-doped quantum wells.[11] Attempts to observe this behavior in the absorption spectrum of GaAs quantum wells in the presence of a high density 2DEG have yielded inconclusive results.[6]

The question that we wish to address in this paper is how does the high-density spectrum transform into the low-density excitonic spectrum? It is well known that at very low densities the absorption spectrum consists of two peaks: one that corresponds to the creation of a neutral exciton X, and another, at a lower energy, that corresponds to the creation of a negatively charged exciton X^-, consisting of two electrons with opposite spins bound to a valence-band hole.[12–14] The X and the X^- are the semiconductor analogs of the hydrogen atom H and hydrogen ion H^-: their spectrum can be fully derived by considering only two- or three-particle interactions, respectively. On the other hand, their final states closely resemble the singly and doubly occupied bound states of the many electron system. A natural question that then arises is whether we can observe any indication in the absorption spectrum that distinguishes between the two regimes.

To address this question we investigate the evolution of the absorption spectrum of a single GaAs/$Al_xGa_{1-x}As$ quantum well with decreasing electron density, from 1.5×10^{11} to 1.5×10^{10} cm^{-2}. We tune the electron density continuously by applying a voltage to a semitransparent gate relative to an Ohmic contact, which is made into the 2DEG. We find that there is a *critical density* that marks the transition from a FES-like to a hydrogenlike behavior. Above this density we observe two peaks with asymmetric line shape. The energy difference between the two peaks, ΔE, increases monotonically with density, and approaches asymptotically the predicted $\Delta E=E_F+E_B$ dependence. Below this critical density the exciton peak becomes symmetric and the energy difference between the two peaks is constant, independent of the density.

The absorption spectrum is obtained using photocurrent spectroscopy. Electrons are excited from surface states by the incident photons to energies above the Schottkey barrier, and then drift into the doped region. From there they tunnel into the well and are collected by the Ohmic contacts of the 2DEG, giving rise to a photocurrent in the gate circuit. This mechanism gives rise to a photocurrent at photon energies that extend well below the GaAs gap. At photon energies that can be absorbed in the quantum well there is an additional contribution to the photocurrent, resulting from electron-hole pairs created in the well. To verify that the measured photocurrent spectrum indeed reflects the absorption spectrum we have conducted complementary photoluminescence excitation measurements. Similar behavior of the peak positions and line shape were observed. There are, however, two important experimental points that should be mentioned.

(1) The photocurrent from the quantum well rides on a spectrally broad background signal, due to surface state absorption. The value of this background photocurrent depends on the sample structure and surface treatment. The spectra are displayed after this background was subtracted.

(2) In this gated structures light illumination gives rise to some depletion of the carrier density. However, since the capacitance of the structure is constant, this only implies that we need to add some positive gate voltage in order to recover the density in the dark. We have verified this behavior over a large light intensity range.

The sample structure that we investigated consists of the following sequence of layers: a 300-nm-thick GaAs buffer, a superlattice with 50 periods of 100-nm $Al_{0.37}Ga_{0.67}As$ and 30-nm GaAs, a 20-nm-thick GaAs quantum well, a 50-nm-thick $Al_{0.37}Ga_{0.67}As$ spacer layer, Si delta-doping of 1×10^{12} cm^{-2}, a 100-nm $Al_{0.37}Ga_{0.67}As$, a 20-nm thick *n*-type $Al_{0.37}Ga_{0.67}As$, and a GaAs cap layer. The wafer is processed into a field-effect transistor structure, with a 5-nm-thick semitransparent PaAu gate. Determination of the electron density N under illumination is done by measuring the photoluminescence (PL) spectrum as a function of magnetic field, and finding the magnetic field values at which an integer number of Landau levels are filled. At these magnetic fields there is an abrupt change in the PL spectrum.[15] This procedure works well up to $N\sim8\times10^{10}$ cm^{-2}. To obtain the lower electron densities we use the capacitance of this structure to extrapolate the curve of $N(V_g)$, where V_g is the gate voltage.

The measurements are conducted in a liquid-helium storage Dewar at a temperature of 4.2 K, using an optical-fiber-based system. We use a single-mode fiber at close proximity, ~100 μm, to illuminate the sample. A second multimode fiber is set at a distance of a few mm from the sample and collects the emitted PL. The sample is excited by a tunable Ti-sapphire laser, which is coupled into the single-mode fiber. The photocurrent is amplified by a sensitive current amplifier and measured using a lock-in amplifier. We have conducted the measurements using power levels from 10 nW to 1 mW. Except for a shift in the $N(V_g)$ curve and some broadening at high power levels, the results are power independent. After an absorption measurement at a given gate voltage V_g, the laser is set to 1.541 eV and the PL is measured.

Figure 1 shows a set of photocurrent and PL spectra taken for various gate voltages. At large electron densities [Fig. 1(a)] the absorption edge is a broad step that is shifted to high energies relative to the PL spectrum. In Fig. 1(b), which is taken at $N=6.6\times10^{10}$ cm^{-2}, we observe the formation of two broad peaks: one at the absorption edge and the other a few meV higher. As the density is further reduced ($N<3.3\times10^{10}$ cm^{-2}) the two broad peaks acquire an asymmetric singular line shape, characterized by a steep rise at low energies and a slow fall at high energies [Figs. 1(c,d)]. Following the notation of Ref. 7 we label the low-energy peak as ω_1 and the high-energy one as ω_2. The singularity in ω_2 increases very fast with decreasing density, and at $N=2.1\times10^{10}$ cm^{-2} it becomes a symmetric resonance, the heavy-hole exciton [Fig. 1(e)]. The ω_1 peak exhibits less pronounced changes. As the density is reduced it becomes weaker and evolves into the well-known charged exciton X^- [Figs. 1(e,f)]. A replica of that scenario appears at the light-hole exciton energy, 4 meV higher.

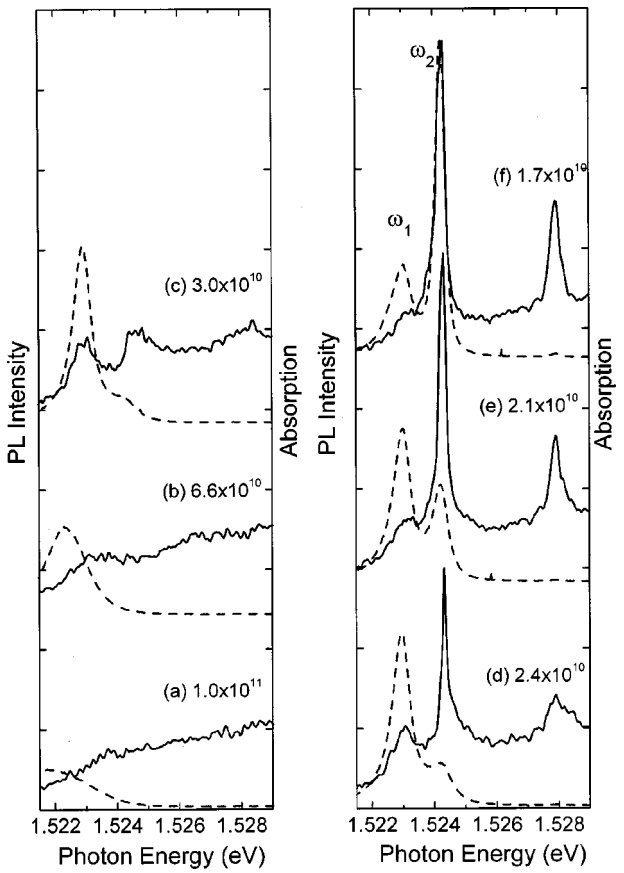

FIG. 1. A series of the absorption (solid line) and PL (dotted) spectra at several gate voltages. Each spectrum is labeled by the corresponding electron density. The spectra are displayed after subtraction of the background signal (see text).

Figure 2 shows the density dependence of the energies of the ω_1 and ω_2 peaks. The data are presented for a limited density range, where the singularity edge is clear and the peak energy can be unambiguously determined. In Fig. 2(a) we show the ω_1 and ω_2 transition energies for both the absorption and PL spectra. It is seen that the ω_2 peak energy is constant over a certain density range and then shifts to higher energies with increasing density. The ω_1 peak energy is also constant at low densities but shifts slightly to lower energies with increasing density. The Stokes shift between the exciton (ω_2) peaks in the absorption and PL spectra is a very small at low densities, <0.1 meV, and increases at higher densities to ~0.2 meV.

In Fig. 2(b) we present the energy difference $\Delta E=\hbar(\omega_2-\omega_1)$ as a function of electron density. It is evident that there exists a critical density value, $N_c\approx2.5\times10^{10}$ cm^{-2}, below which $\hbar(\omega_2-\omega_1)$ is constant and equals 1.2 meV. Only above this critical density the energy difference $\hbar(\omega_2-\omega_1)$ grows, first rapidly and then more gradually. The dashed line describes the relation $\hbar(\omega_2-\omega_1)=E_B+E_F$ (where $E_B=1.2$ meV). It is evident that the measured points in Fig. 2(b) are below this curve at low density, and as the density gets higher the data points approach the dashed line.[16]

It is remarkable that the transition is very sharp and is seen both in the photocurrent and PL spectra, though in

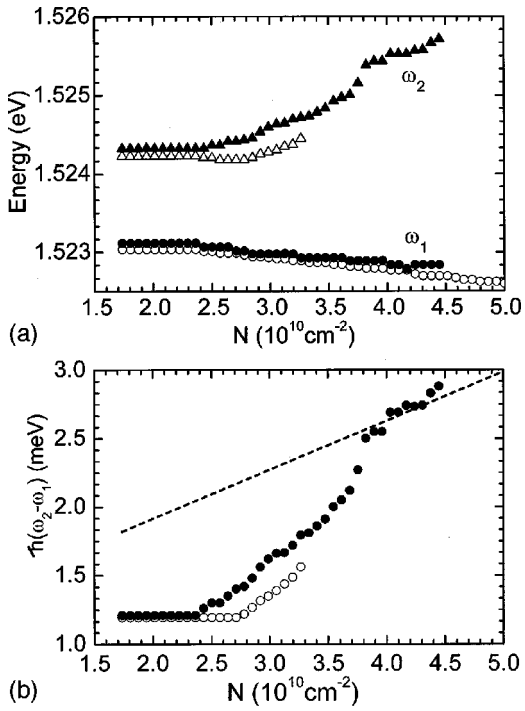

FIG. 2. (a) The energies of the ω_1 and ω_2 transitions as a function of electron density. The solid and empty symbols represent the peaks position in absorption and PL, respectively. (b) The energy difference $\hbar(\omega_2-\omega_1)$ as a function of electron density in the absorption (solid circles) and PL (empty circles) spectra. The dashed line describes the relation $\hbar(\omega_2-\omega_1)=E_B+E_F$, with $E_B=1.2$ meV.

slightly different densities, 2.5×10^{10} cm^{-2} and 2.9×10^{10} cm^{-2}, respectively. It should be emphasized that the optical spectra [Figs. 1(e,f)] clearly show that the density is indeed changing below N_c, as evidenced by the exchange of oscillator strength between the X and the X^-. This conclusion is supported by transport measurements through the sample at this range, which show that the conductivity decreases as the gate voltage becomes more negative. The observed behavior is in clear contrast with that reported in Ref. 11, where it was argued that $\hbar(\omega_2-\omega_1)$ changes linearly with E_F all the way to zero density. In fact, if we would have deduce the zero density X^- binding energy by extrapolating the high density behavior as done in Ref. 11, we would have obtained a ridiculously low value of $\sim$0.4 meV. This value is in clear disagreement with theoretical calculations and ultralow density PL measurements, which sets the binding energy for this well width at approximately $\sim$1 meV.

A close examination of Fig. 1 reveals that at the critical density N_c not only the energy of the ω_2 peak shifts but also its line shape undergoes a drastic change: it can be seen that is symmetric below N_c [Figs. 1(e,f)] and is a singular asymmetric line above it [Fig. 1(d)]. As the density is increased the slope of the high-energy tail becomes smaller [Fig. 1(c)]. The critical density appears to be distinguishing between two regimes: a high density regime, in which the exciton energy and line shape is sensitive to the presence of a Fermi sea, and a low density regime where the exciton behavior is of a hydrogenlike object. The question that arises is what defines the density scale for the problem. One possible candidate is the X^- area, S_{X^-}. We note that when the density is lower

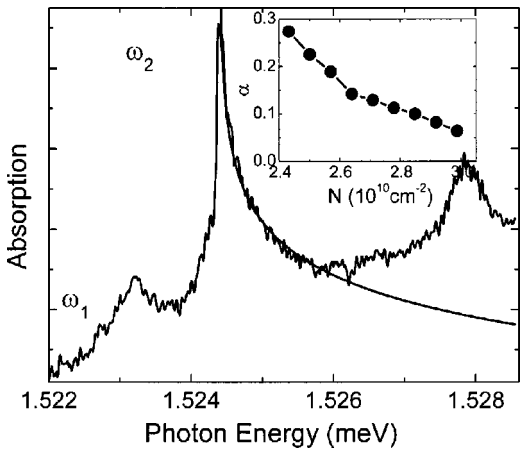

FIG. 3. (a) A fit of the function $A(\omega)=(\omega-\omega_0)^{-\alpha}$ to the exciton (ω_2) singularity at $N=2.7\times10^{10}$ cm^{-2}. The inset shows the dependence of the exponent α on electron density.

than $1/S_{X^-}$ the X^- bound state is formed by the photoexcited electron and one of electrons of the 2DEG. As a result, this area is depleted of electrons, and there is no cost of E_F energy to pay in ionizing this state. Hence, the second peak is formed at an energy E_B above the first one and not at E_B+E_F. This argument also explains the change of the exciton line shape around N_c. Only when a Fermi sea of electrons is present within this area and shake-up processes can occur, a high-energy tail is formed. Indeed, the observed critical density agrees with the estimated size of X^-, which has a radius of $\sim$30 nm. Another physical quantity that defines a density scale is the temperature. We note that at the critical density $E_F\approx2.5$ kT, and consequently there are available states below E_F, which can participate in the X-X^- absorption. This could explain why $\hbar(\omega_2-\omega_1)$ is independent of density below N_c. We find it difficult, however, to apply this argument to explain the changes in line shape.

The apparent insensitivity of the optical spectrum to the background electron density below N_c is observed also in the behavior of the shake-up satellites lines in the PL spectrum at low magnetic fields. These satellite lines are due to a recombination process in which an electron from the 2DEG is ejected to a high Landau level.[17] We find that below N_c the energy of the first shake-up line, SU_1, which corresponds to a final state where the electron is in the first Landau level, is at $\hbar\omega_c$ below the X^- line (where ω_c is the cyclotron frequency). On the other-hand, above N_c the shake-up broadens and shifts to lower energies, corresponding to excitation of the 2DEG by an energy larger than $\hbar\omega_c$. It was previously shown that this excess energy is due to the creation of magnetoplasmons, a collective excitation of the 2DEG.[18]

An important issue to be considered in this context is the role of electron localization at the very low density regime. We have previously shown that the appearance of excitons in the PL spectrum marks the onset of strong localization of the electrons in the potential fluctuations of the ionized donors. These donors, which are at a distance of 50 nm (the spacer width) in our sample, are randomly distributed in the plane and induce a spatially fluctuating electrostatic potential at the 2DEG. At high electron density the 2DEG efficiently screens the fluctuations, but as the density is reduced the screening becomes less efficient and they grow considerably.[19] At a

certain density this process gives rise to localization of the electrons. This density range, in which the screening capability of the localized electrons depends on their density, is usually called the nonlinear screening regime. Indeed, near-field PL measurements have shown that in this regime the intensity of the X^- is very inhomogeneous and strongly fluctuates from one location to another, providing a direct evidence for the electron localization.[20] We shall show below that the disorder potential plays an important role in determining the absorption spectrum.

We notice that the evolution of the singularity above N_c is surprisingly fast. Figure 3 describes a fit of a power-law singularity $A(\omega)=(\omega-\omega_0)^{-\alpha}$ to the high-energy tail of ω_2 at $N=2.7\times10^{10}$ cm^{-2}. Such a fitting procedure allows us to accurately extract the exponent α at each density. The inset shows the dependence of α on N. It is seen that α decreases by substantially (from 0.3 to 0.05) in a very narrow range, $\Delta N=8\times10^9$ cm^{-2}, which corresponds to increasing the Fermi energy by less than 0.25 meV. We find that α diminishes at the density at which the exciton line disappears from the PL spectrum. The exponent α is related to the phase shift of the electrons at the Fermi surface, when scattering off the valence hole potential.[9] In that sense it measures the efficiency of the Fermi sea electrons in screening that potential: the smaller α, the better the screening. Since in the nonlinear screening regime the screening capability depends on the density we expect the exponent α to vary as well. It should be noticed that our sample is of relative high quality. This is evidenced by the high mobility ($\sim10^6$ cm^2 V^{-1} s^{-1}), the narrow exciton linewidth (0.3 meV), and the small Stokes shift between the PL and absorption (<0.1 meV). Thus, at $N>3\times10^{10}$ cm^{-2}, where the potential fluctuations are suppressed, our sample could be considered as close to ideal, and the singularity disappears. Only when disorder sets in, and electrons cannot efficiently screen the valence-hole potential, α becomes large enough to make ω_2 and ω_1 observable as singular peaks.

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