Evolution of the interband absorption threshold with the density of a two-dimensional electron gas

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We have obtained absolute interband absorption spectra from a series of modulation-doped GaAs/$Al_{0.3}Ga_{0.7}As$ multiple quantum wells covering a wide range of two-dimensional electron densities. The evolution of the threshold line shape from an exciton to a Fermi-edge singularity is quantified. We discuss the influence of the doubly occupied bound state (X^-) on the line shape and compare the results with realistic many-body calculations.

Interband optical absorption spectra from undoped, direct-gap semiconductor heterostructures are characterized by two thresholds: a discrete excitonic feature due to excitation of a bound electron-hole pair, and a well-defined steplike continuum edge shifted up in energy from the bound state by the exciton binding energy, E_b.[1] In contrast, in the presence of a high density of free carriers the spectra exhibit only a single threshold that typically appears as a steplike edge enhanced at the threshold. The enhancement in the vicinity of the Fermi level [the Fermi-edge singularity (FES)] (Refs. 2–7) is similar to that observed in x-ray absorption spectra of metals, and results from the many-body response of the mobile two-dimensional electrons to the photoexcited valence hole.[1–9] Despite intensive investigations of the optical properties of semiconductor heterostructures, the evolution between these two extremes, as a function of the density n_s (or Fermi energy E_F) of a two-dimensional electron system (2DES), is not well understood.

This problem was addressed theoretically by Combescot and Nozieres[9] (CN) for the 3D metallic case. They found that at all densities the excitonic and continuum thresholds have absorption line shapes (for $\omega-\omega_i \ll E_F$) given by the power-law singularity

$$A_i(\omega)=1/(\omega-\omega_i)^{\alpha_i}. \tag{1}$$

CN showed that as n_s increases the sharp excitonic feature should broaden asymmetrically to higher energies ($1 \gtrsim \alpha \gtrsim 0$) and the steplike continuum edge should become increasingly rounded ($0 \gtrsim \alpha \gtrsim -3$), with the effects most significant for $E_F \gtrsim E_b$.

Recent experimental work[10–12] has shown that in the case of semiconductor heterostructures containing a low excess 2D carrier density, an exciton can bind an extra electron causing the formation of a negatively charged exciton X^-. The X^- feature appears $\sim$1 meV to lower energies than the exciton. The potential of the valence hole thus supports a *doubly* occupied bound state, which should continue to exist even in the presence of a dense 2D electron gas.[13] Hence, *a priori*, there should always be *three* thresholds in the absorption spectrum, ω_1, ω_2, and ω_3 (in order of increasing energy), corresponding to occupancy of the bound state by two, one, and zero electrons. Physically, these transitions correspond to excitation of an electron into X^- (the ground state, with the second electron provided by the Fermi sea), the exciton, and the continuum of unbound states (see Ref. 14). The actual line shape for each threshold, as a function of carrier density, must be determined and understood by suitable calculations, and by experiment.

The CN formalism is not directly applicable to absorption in the presence of a 2DES because it only considers a single spin component of the electrons, ignores electron-electron interactions, and assumes an infinite hole mass. In the 2D case (i) different spin channels cannot be treated as independent because of the existence of the doubly occupied bound state, X^-, (ii) electron–electron interactions must be included because of the presence of X^- and because of the fact that 2DES's exhibit gapless plasmon excitations, and (iii) valence holes in semiconductor heterostructures are mobile, with effective masses of the order of the free electron mass.

The CN approach has previously been developed by one of us[14] into a realistic model of the 2D case that considers the evolution of the spectra from the excitonic limit to the FES. The model includes the doubly occupied bound state, and treats electron-electron interactions within the random phase approximation (RPA). Numerically generated absorption spectra[14] reveal a fundamental (ω_1) threshold that broadens

asymmetrically with increasing n_s (similar to CN for the threshold associated with the exciton), and a strong *peak* at ω_2. The separation $\omega_2-\omega_1$ represents the minimum energy required to remove one of the bound electrons from the ground state (X^-) in the presence of a valence hole. There is no feature in the calculated spectrum at the continuum (ω_3) threshold.

Previous optical experiments[3–7,15] on modulation-doped semiconductor systems have clearly established that the absorption threshold exhibits an asymmetric enhancement, qualitatively similar to the FES observed in metals, for $n_s \gtrsim 2\times10^{11}$ cm^{-2} ($E_F \gtrsim 7$ meV). The enhancement becomes less pronounced as n_s increases at a fixed temperature, or if the temperature is increased at a fixed density. Existing experiments[5,15] that to some extent address the issue of how the zero-density exciton evolves into a FES suggest that there is a continuous evolution between the two extremes, but the samples and/or techniques used in these studies preclude a quantitative, or even semiquantitative interpretation. Of particular interest are the relative energy scale E_F/E_b over which this transition occurs, quantification of the strength of the singularity as a function of carrier density, and an understanding of the role of X^-.

In this paper we present absolute absorption spectra that allow us to quantify the evolution of the threshold line shape as the density of the 2DES is varied between $n_s=0$ and $n_s=1.4\times10^{12}$ cm^{-2}. From a comparison of these spectra with numerical calculations, and from their temperature dependence, we conclude that for $n_s>0$ the dominant threshold is consistent with optical excitation of the ground state of the many-electron system in the presence of the valence hole, i.e., the state with a doubly occupied bound state. As the density increases, the influence of this bound state on the threshold *line shape* decreases dramatically even for $E_F \ll E_b$ (i.e., for carrier densities where only a small fraction of an electron from an uncorrelated Fermi sea would be within the Bohr radius of the bare exciton), consistent with theoretical predictions of a dramatic reduction in the bound state binding energy at finite n_s (E_b').

The samples investigated are 50-period modulation-doped 50-Å GaAs/400-Å $Al_{0.3}Ga_{0.7}As$ multiple quantum wells (MQW's) with electron densities ranging from $n_s=0$ to $n_s=2\times10^{11}$ cm^{-2} and a pair of 50-period 60-Å MQW samples with $n_s=7\times10^{11}$ and 14×10^{11} cm^{-2}. The samples were removed from their substrates using an epitaxial lift-off technique and subsequently Van der Waals bonded to glass substrates to allow absorption measurements.[16] Absorption was measured using broad-band, polarized light incident near Brewster's angle.

Figure 1(a) shows the absolute absorption spectra of the 50-Å samples at 5 K in the vicinity of the threshold. The $n_s=0$ sample (top) has strong heavy hole (hh) and light hole (lh) exciton peaks at 1.620 and 1.6475 eV, as well as a clear hh-continuum edge at 1.632 eV and a weaker lh-continuum edge at ~1.660 eV. The hh and lh exciton binding energies (E_b) are therefore both ~12 meV. Figure 1(b) shows absorption spectra for the higher-density 60-Å samples. The fits to the hh line shape (dashed lines) are discussed below.

Figure 2 shows the energy of the absorption edge (half-intensity point on the rising edge) and the corresponding photoluminescence peak energy for the 50-Å samples. The

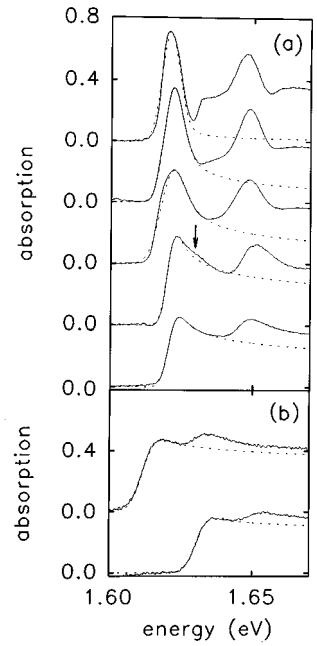

FIG. 1. Absorption spectra for (a) the 50-Å MQW samples with $n_s=0$, 0.25, 0.5, 1.0, and 2.0×10^{11} cm^{-2} (from top to bottom) and (b) the 60-Å MQW samples with $n_s=7$, 14×10^{11} cm^{-2}. The fits to the absorption threshold (dashed line) are discussed in the text.

Stokes shift for each sample is in good agreement with that expected from the nominal doping densities: the actual 2D densities are therefore close to the nominal values. Figure 2 also demonstrates that differences in well width from sample to sample are not significant.

In the remainder of this paper we discuss the results for the hh features (the lh features show qualitatively similar behavior). We begin by pointing out that the threshold line shape is dramatically modified by the presence of a remarkably low density of free carriers [$n_s\sim2.5\times10^{10}$ cm^{-2}; see Fig. 1(a)]. Even in the lowest density doped sample, only a single threshold is observed, and it clearly emanates from the exciton peak at zero density, not from the continuum edge.

In addition, the temperature dependence (not shown) of the fundamental threshold of samples with large n_s ($\gtrsim2\times10^{11}$ cm^{-2}) is characteristic[2–6] of the FES, i.e., the peak is eliminated by raising the temperature to ~50 K. In the undoped sample the excitonic feature is virtually temperature independent up to 100 K. For intermediate densities the en-

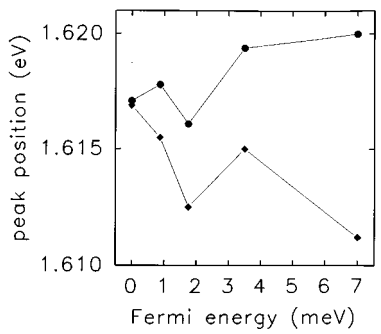

FIG. 2. Positions of the photoluminescence peak (diamonds) and the half intensity point on the rising edge of the absorption peak (circles).

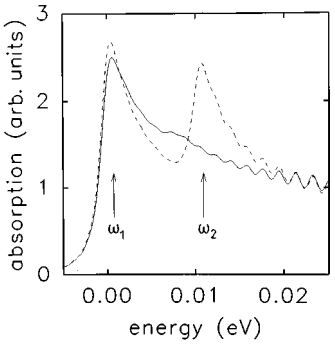

FIG. 3. Calculated absorption line shape for $n_s = 1.8 \times 10^{11}$ cm^{-2} in RPA (dashed line) and in a self-consistent Hartree approximation (full line). Features in the RPA spectrum corresponding to double (ω_1) and single (ω_2) occupancy of the bound state are labeled. The weak oscillatory structure comes from the use of a discrete set of energy levels.

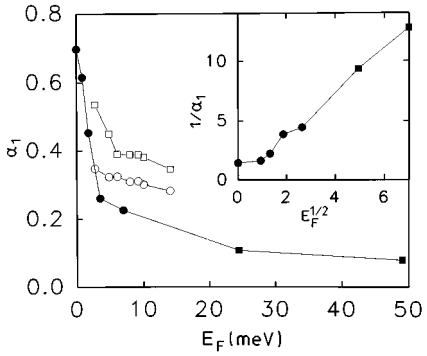

FIG. 4. Exponents α_1 from the fits to the experimental data of Fig. 1 using the line shape of Eq. (1): filled circles, 50-Å wells; filled squares, 60-Å wells. Also shown are the calculated values of α_1: open squares, RPA; open circles, Hartree approximation. The inset shows that $1/\alpha_1$ depends linearly on Fermi wave vector ($\sim E_F^{1/2}$) for large E_F.

ergy, line shape, and temperature dependence of the threshold all show that as n_s increases, there is a continuous evolution toward the FES as many-body effects become more dominant and the bound state more strongly screened.

The prediction[14] of two peaks in the spectrum (at ω_1 and ω_2) is clearly not verified by experiment. The only experimental evidence for an additional (ω_2) peak is the weak feature in the spectrum of the $n_s = 1 \times 10^{11}$ cm^{-2} sample (arrowed in Fig. 1). We note that for samples with lower densities the calculated separation of the peaks, $\omega_2 - \omega_1$,[14] is smaller than the inhomogeneous broadening in our samples, making it impossible to resolve such a weak feature.[17]

The presence of only a single threshold suggests that it is necessary to go beyond the theory of Ref. 14 to properly describe the experimental spectra. A formalism has recently been developed[18] that allows treatment of electron-electron interactions within either the RPA or a self-consistent Hartree approximation. This formalism can be used to calculate the absorption line shape for a limited range of carrier densities ($n_s = 0.7 - 4.0 \times 10^{11}$ cm^{-2}) for the case of localized holes. We compare results from the RPA and Hartree approximations for $n_s = 1.8 \times 10^{11}$ cm^{-2} in Fig. 3. The shape of the first threshold, corresponding to creation of the X^- ground state, is relatively insensitive to the treatment of electron-electron interactions but the ω_2 peak is dramatically weakened in the Hartree calculation, yielding a feature similar to that arrowed in Fig. 1. Details of these calculations will be reported elsewhere.

The new line-shape calculations allow a direct comparison of experiment and theory. The line shapes of the experimentally observed and *numerically* calculated singularities can be fitted using the functional form of Eq. (1): the fitted exponent α_1 provides a phenomenological measure of the strength of the singularities. Our procedure (following that of Ref. 19 for the 3D metallic case) leads to good fits to the experimental data (dashed lines in Fig. 1) over essentially the whole energy range of the hh peak for all samples. In this fitting procedure we find that Lorentzian lifetime broadening may be ignored but convolution with a Gaussian is important: in this case it likely represents inhomogeneous broadening rather than instrumental or phonon broadening effects.[19] Note that while a number of free parameters (e.g., line position, peak height) are used in the fits the only parameters which affect the line shape are the width of the Gaussian broadening, γ,[20] (a relatively small effect) and the exponent α_1. The temperature is accounted for by a convolution with the derivative of the Fermi function at a fixed temperature (5 K). Figure 4 shows that the Hartree calculation produces line shapes that are reasonably close to the experimental ones. The RPA line shapes are narrower (α_1 larger) because the RPA neglects the repulsion between the two bound electrons and therefore overestimates the strength of the bound state. This comparison of experimental and theoretical absorption line shapes leads us to conclude that although, in principle, one could expect three distinct thresholds, in the presence of a dense 2DES there is essentially only a single threshold corresponding to excitation of the ground state, which includes the doubly occupied bound state X^-.

α_1 is a measure of how much of the total screening of the valence hole is done by the bound electron generated in the absorption process, as compared to the screening provided by rearranging electrons in the Fermi sea.[9] At zero density the bound state electron does all the necessary screening, and for $E_F \gg E_b$ it is intuitively clear that several of the electrons from the Fermi sea will be within the radius of the bound state, and therefore they will provide the bulk of the screening despite the presence of the bound state. This suggests[9] that the spectrum should remain excitonlike (X^--like in the 2D case), with a large contribution from the bound state, until $E_F \sim E_b \sim 12$ meV. The Hartree calculation, however, shows that the binding energy of the doubly occupied bound state is small and nearly constant ($E_b' \sim 1.7$ meV *per electron*) for the density range over which the calculation is valid ($E_F = 3 - 14$ meV). This dramatic decrease in binding energy indicates that screening by the bound electrons is much reduced and that the Fermi sea plays an important role in screening the valence hole even for $E_F < 3$ meV, hence the rapid reduction in the experimental value of α_1 as carriers are introduced.

These results show that the influence of the doubly occupied bound state on the threshold line shape at high n_s is rather subtle. At high densities the bound state is responsible for a small fraction of the total screening of the valence hole and so the value of α_1 is small (as in the 3D metallic case—

compare Refs. 8 and 9). The feature of the data that might provide a definitive test of the role of the doubly occupied bound state at high n_s is the density dependence of the line shape [and particularly the slope of the plot of $1/\alpha_1$ versus $E_F^{1/2}(\sim k_F)$; see Fig. 4 (inset)]. A theoretical challenge is to develop a formalism which allows an accurate treatment of interactions over a wide range of n_s, while accounting for the finite mass of the valence hole in a realistic way. The fact that the finite mass of the valence hole does not seem to influence the qualitative nature of the experimental absorption spectra is an outstanding theoretical problem.[14,21]

In summary the comparison of the experimental absorption line shapes with the new RPA and Hartree calculations yields the following main points: (i) the strong additional ω_2 peak in the RPA calculations, associated with creation of a neutral exciton in the 2DES is found to be very much weaker when interactions are treated in a self-consistent Hartree approximation, consistent with the observation of a single threshold in the experiment. (ii) Many-body effects modify the threshold line shape at remarkably low carrier concentrations: it broadens much more quickly than expected in the[9] CN and[14] RPA treatments of the problem. (iii) The experimental and calculated threshold line shapes can be characterized by a single power-law exponent (α_1), over the whole range of densities studied. (iv) The Hartree line shapes agree reasonably well with the measured line shapes over the range of densities where the calculation is reliable, whereas RPA calculations do not describe the data.

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