Excitonic Recombination of Degenerate Two-Dimensional Electrons with Localized Photoexcited Holes in a Single Heterojunction Quantum Well

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We report the observation of the excitonic recombination of degenerate quasi-two-dimensional electrons with localized photoexcited holes. Low-temperature photoluminescence and magnetophotoluminescence spectra exhibit a well resolved Mahan exciton resonance which is sensitive to electron density n_s. We observe a sharp decrease in the exciton oscillator strength and linewidth with the concomitant formation of biexcitons and a large discontinuity in the exciton ground-state energy at $n_s \approx 1.9 \times 10^{11}$ cm^{-2}. An abrupt transition from excitonic to free-electron–localized-hole recombination occurs at $n_s \approx 2.2 \times 10^{11}$ cm^{-2}.

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In a seminal paper [1] Mahan pointed out that at low temperatures stable electron-hole bound states, Mahan excitons, can exist in degenerate semiconductors whose carrier density is lower than an unspecified limit. He also predicted the existence of the Fermi edge singularity (FES) which is caused by multiple electron-hole scattering near the Fermi edge of a degenerate electron gas. The recent two-dimensional (2D) theory [2] and observation of the many-body FES in the low-temperature photoluminescence [3] and absorption [4,5] spectra of semiconductor modulation-doped quantum wells (QW) has generated significant research interest [6,7]. If it exists, the lowest Mahan exciton state would appear in the photoluminescence (PL) or absorption spectrum of the degenerate electron gas at the energy [1] $E_{en} = E_g + p_F^2/2v - E_n$ where E_g is the energy gap, E_n is the binding energy, p_F is the electron Fermi momentum, v is the reduced mass, and the Fermi edge occurs at $E_g + p_F^2/2v$. It is expected that in the presence of a Fermi gas the Mahan exciton eigenstates will depend on various many-body interactions and effects [1,8–15]. To our knowledge, no definitive observation of excitonic recombination in the PL spectrum of a degenerate electron gas has been reported. Spatially indirect radiative recombination of electrons with free [7] or bound [16,17] photoexcited holes in an AlGaAs/GaAs single heterojunction quantum well (SHQW) has been observed before; however, in these cases there is either no evidence [16,17] or slightly suggestive evidence [7] of excitonic recombination. In this paper we report the first observation of the Mahan exciton in the low-temperature PL spectrum arising from the recombination of bound and free-electron–localized-hole (e-h) pairs in the presence of a two-dimensional electron gas (2DEG) in a SHQW whose electron density n_s is varied over the range $1.3 \times 10^{11} \le n_s \le 2.3 \times 10^{11}$ cm^{-2}.

The experiments are carried out on a modulation-doped coupled well heterostructure which consists of the following layers: a semi-insulating GaAs substrate, a 3000 Å undoped GaAs buffer layer, 50 Å undoped $Al_{0.3}Ga_{0.7}As$, 50 Å n-type GaAs (2×10^{18} cm^{-3} Si doping), 800 Å $Al_{0.3}Ga_{0.7}As$ (1×10^{18} cm^{-3} Si doping), and 100 Å GaAs (1×10^{18} cm^{-3} Si doping). Shubnikov–de Haas measurements at 4.2 K on the unilluminated sample show that the electrons are confined only in the lowest subband of the SHQW formed by the transfer of electrons from the doped AlGaAs into the undoped GaAs buffer region. Without illumination the measured 2D electron density and mobility are 2.9×10^{11} cm^{-2} and 5×10^4 cm^2/V s, respectively. n_s was varied by applying a voltage between an Ohmic contact to the 2DEG and a Schottky contact to the 120-μm-thick substrate; see Fig. 1. The samples were mounted in a variable temperature liquid helium cryostat and excited with the 5145 Å line from an argon ion laser with an intensity of ~ 1 W/cm^2. The luminescence spectrum was measured with a resolution of 0.1 meV using a 1 m monochromator. An optical fiber apparatus was used for the magneto-PL measurements with the sample mounted in the bore of a superconducting magnet.

The observed 6 K PL spectrum with the substrate voltage $V_g = 0$ shows the typical emission from the GaAs substrate [18] and the doped AlGaAs layer. In addition, we observe a 27-meV-wide (ionized impurity scattering broadened [19]) luminescence signal from the doped QW at 1605 meV and an unusual signal, shown in Fig. 2(a), which has a steep increase in intensity at ~ 1556.5 meV and a sharp peak at 1553.3 meV. As n_s is reduced, the intensity of the 1553.3 meV resonance increases slightly over the range -10 V $\le V_g \le 0$ and then stays constant

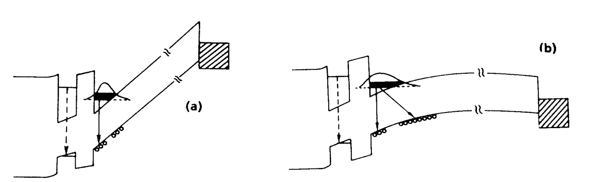

FIG. 1. Schematic of the backgated coupled well band structure for (a) negative substrate bias and (b) positive substrate bias.

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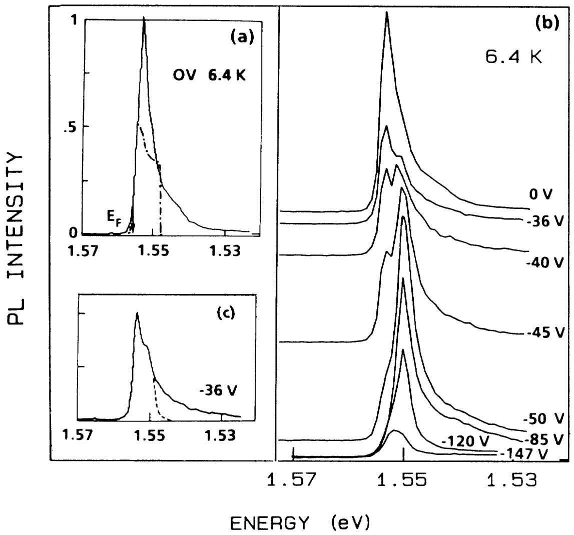

FIG. 2. The observed 6.4 K PL spectrum from the SHQW for various substrate voltages. Some of the spectra shown in (b) have been arbitrarily shifted along the vertical axis to enhance visual clarity. The dot-dashed line in (a) is the calculated Fermi edge enhancement. The approximate location of E_F in (a) is obtained from the analysis of PL data at $V_g = 4$ V. The dashed line in (c) is an extrapolation of the low-energy cutoff.

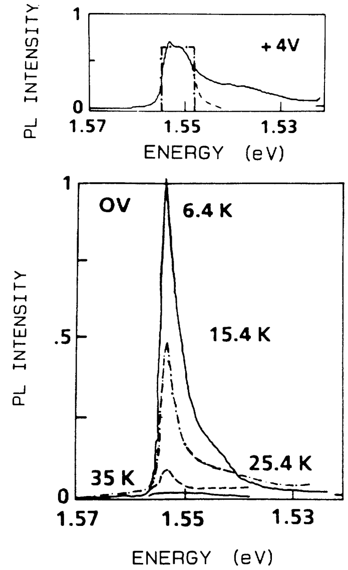

FIG. 3. (a) The observed 6.4 K PL spectrum from the SHQW for $V_g = 0$ is shown by the solid line. The dot-dashed line shows the theoretical PL line shape for free-2D-electron–localized-hole recombination, $T = 0$, $n_s = 2.2 \times 10^{11}$ cm^{-2}. The dashed line is the extrapolation of the low-energy cutoff. (b) Observed PL spectra for various temperatures with $V_g = 0$.

as V_g varies from -10 to -30 V. As n_s is further reduced the amplitude and linewidth of the 1553.3 meV resonance decrease abruptly, allowing the clear resolution of a shoulder in the PL spectrum at 1550.4 meV as shown in Figs. 2(b) and 2(c) for $V_g = -36$ V. For -40.5 V $\leq V_g \leq -39$ V we observe, as shown in Fig. 2(b) for $V_g = -40$ V, the reduced-amplitude 1553.3 meV resonance and another peak at 1551.5 meV whose amplitude $\propto I^{1.4}$, where I is the laser intensity. At $V_g = -41$ V, the sharp 1551.5 meV peak abruptly disappears and at $V_g = -42$ V we observe a sharp peak at 1550.2 meV in addition to the 1553.3 meV peak. The intensities of both peaks are equal. Over the narrow range -47 V $\leq V_g \leq -42$ V both the 1553.3 and 1550.2 meV peaks are observed; the PL intensity at 1550.2 meV increases rapidly while the intensity at 1553.3 meV decreases slowly over this range. For $V_g < -47$ V we do not observe the 1553.3 meV resonance. In contrast, the 1550.2 meV resonance increases sharply as n_s decreases, attains a maximum intensity at $V_g = -50$ V, remains constant over the range -70 V $\leq V_g \leq -50$ V, and then gradually decreases leaving a small, relatively broad feature at $V_g = -147$ V, as shown in Fig. 2(b). No PL tail (energy < 1540 meV) is observed for $V_g \leq -120$ V. The sharp 1553.3 meV resonance which is observed at $V_g = 0$ is abruptly eliminated by slightly increasing n_s by setting $V_g \geq 4$ V as shown in Fig. 3(a); the PL exhibits the steep increase in intensity at ~ 1556.5 meV, a region where the intensity is roughly constant and a low-energy tail which is about 8 meV longer than that at $V_g = 0$. The sensitivity of the PL over the range 1556–1540 meV to n_s suggests that this PL comes from the recombination of equilibrium electrons in the SHQW with photoexcited holes which must be localized at the AlGaAs/GaAs interface in the SHQW. The dependence of the low-energy tail on V_g indicates that it can be attributed to the spatially indirect recombination of SHQW electrons with photoexcited holes which are localized approximately 100–150 Å from the interface. The observed SHQW PL linewidths are narrower than comparably modulation-doped QW PL linewidths which are mainly inhomogeneously broadened by the growth-intrinsic fluctuations in the QW size.

If many-body effects are negligible, the PL from the recombination of free 2D electrons in a QW with localized photoexcited holes at temperature $T = 0$ has a square pulse line shape whose width $\approx E_F$, the electron Fermi energy relative to the bottom of the subband [3]. Excluding the low-energy tail, the observed 6 K PL for $V_g \geq 4$ V can be approximated by a square pulse line shape as shown in Fig. 3(a). For spatially direct recombination of SHQW electrons at the Fermi surface with holes that are weakly bound at the interface with energy E_h, the expected emission energy $\hbar\omega_F = E_g - \Delta E_g - E_h + e_0 + E_F$, where e_0 is the energy of the bottom of the SHQW ground-state subband and ΔE_g is the band-gap renormalization. Figure 3(b) shows that no PL from the SHQW is observed for $T \geq 32$ K, indicating that $E_h \approx 2.5$ meV. For $n_s \approx 2 \times 10^{11}$ cm^{-2} the calculated [20] values for e_0, E_F, and ΔE_g are 36, 8, and 6 meV, respectively. Using

$E_g = 1.519$ eV and the above parameters we obtain $\hbar\omega_F = 1.5545$ eV which agrees with the approximate value of 1.555 eV obtained from the data shown in Fig. 3(a) by estimating the Fermi edge of the SHQW 2DEG as the energy at which the PL intensity equals half its intensity at the shoulder. This analysis strongly indicates that the Fermi surface of the SHQW 2DEG is observed as a sharp cutoff on the high-energy side of the PL spectra. E_F is approximately given by the full width at half maximum (FWHM) of the pulse obtained by extrapolating the low-energy cutoff of the PL data shown in Fig. 3(a). Using $E_F = n_s h^2/4\pi m^*$, where h is Planck's constant, the GaAs electron effective mass $m^* = 0.068 m_0$, and m_0 is the electron rest mass, we obtain $n_s \approx 2.2 \times 10^{11}$ cm^{-2} for $V_g = 4$ V. The PL data at $V_g = -147$ V and other data which clearly exhibit a low-energy shoulder were analyzed to obtain approximate values for n_s and E_F with an estimated uncertainty of $\pm 10\%$. We find that n_s varies linearly with V_g with a depletion rate $dn_s/dV_g \approx 6.3 \times 10^8$ cm^{-2}V^{-1}.

In order to confirm that the PL of interest comes from the radiative recombination of SHQW electrons with localized photoexcited holes, PL spectra were measured with a magnetic field H, normal to the QW plane of another sample from the same wafer at 4.2 K with $V_g = 0$. Figure 4(a) shows the observed spectra at $H = 0$ and $H = 3$ T. At $H = 3$ T we resolve a peak at a slightly higher energy than the $H = 0$ resonance and two other peaks which can only be attributed to the recombination of SHQW electrons in the occupied $n = 0$ and $n = 1$ Landau levels with localized holes. The peaks in the low-energy tail are due to the spatially indirect recombination of SHQW electrons with holes which are localized 100–150 Å from the interface. Figure 4(b) shows that the resonance seen at $H = 0$ exhibits a diamagnetic shift $\Delta E \approx 0.034 H^2$ meV over the range $0 \leq H \leq 9$ T. This diamagnetic shift is smaller than that reported for GaAs QW [21,22] and bulk [23] free excitons. These data confirm that the zero-field PL resonance comes from the recombination of Mahan excitons. Figure 4(b) shows that the $n = 1$ and $n = 0$ peak energies increase linearly with H for $H \leq 2.5$ T and $H \leq 4.5$ T, respectively, confirming that for $H = 0$, free e-h recombination contributes to the observed PL for $V_g = 0$. The observed shifts of the $n = 1$ and $n = 0$ Landau levels are smaller than expected. Over the respective ranges $2.5\ \mathrm{T} < H \leq 5.5$ T and $5.5\ \mathrm{T} \leq H \leq 9$ T the $n = 1$ and $n = 0$ PL peak energies increase with a H^2 dependence but remain roughly 0.9 meV below the Mahan exciton energy indicating the formation of magnetoexcitons possibly as a result of the hybridization of the Mahan exciton and the $n = 1$ and $n = 0$ Landau levels, respectively. The maximum PL intensities in both ranges are roughly 40% greater than the peak intensity at $H = 0$. The $n = 1$ Landau level transition is not observed for $H > 5.5$ T. At $H = 5.6$ T the PL peak intensity abruptly decreases by 30% and concurrently an increase in the $n = 0$ transition energy is observed; see Fig. 4(b). These data agree with previous observations [24] and the theoretical prediction that an increase in the PL energy occurs when the highest occupied Landau level is full [25].

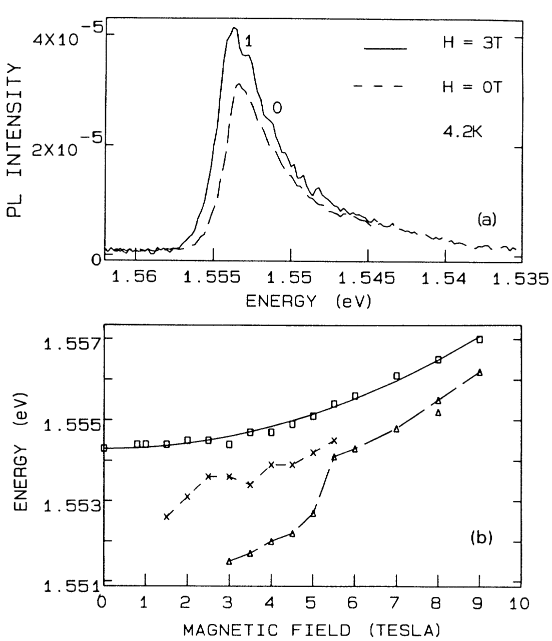

FIG. 4. (a) Observed 4.2 K PL spectra for $H = 0$ T (dashed line) and $H = 3$ T (solid line) with $V_g = 0$. The marked peaks are the $n = 0$ and $n = 1$ SHQW Landau level transitions. (b) Observed Mahan exciton (squares), the $n = 1$ (crosses) and $n = 0$ (triangles) transition energies as a function of the magnetic field. The solid line is a least squares fit to the data; the dashed lines are visual guides.

Following a previous treatment [26] we calculated the Fermi edge enhancement due to multiple e-h scattering without vertex corrections using the Fang-Howard variational wave function [20] to account for the quasi-2D nature of the electron confinement in the SHQW for the case where $n_s = 2.2 \times 10^{11}$ cm^{-2}, $T = 6.4$ K, and the hole mass is infinite. The resulting PL intensity shown by the dot-dashed line in Fig. 2(a) shows that the Fermi edge enhancement (defined as the ratio of the peak intensity to the intensity at e_0) is 1.6, which is consistent with a similar calculation for a QW [27]. This enhancement would be reduced to 1 if hole lifetime broadening [1] and the finite hole mass [28] were accounted for and it is not sensitive to small changes in n_s. On this basis we conclude that the observed PL resonance is not the Fermi edge singularity.

These results verify that, for $V_g = 0$, we are observing the superposition of the emission from the recombination of free and bound e-h pairs in the SHQW. The Mahan exciton ground-state emission results in a strong resonant enhancement of the PL intensity at E_{en}. Figure 3(a) explicitly shows that an abrupt transition from excitonic to free e-h recombination occurs at $n_s \approx 2.2 \times 10^{11}$ cm^{-2}

($V_g = 4$ V) due to many-body effects [1,8–15]. The abrupt nature of the unbinding of the Mahan excitons suggests that phase space filling plays a key role [10,11]. Over the range 1.9×10^{11} cm^{-2} < n_s < 2.2×10^{11} cm^{-2} the Mahan exciton ground-state energy and binding energy are 1553.3 and 1.7 meV, respectively. For n_s slightly above 1.9×10^{11} cm^{-2} ($V_g = -40$ V), Fig. 2(b) shows that the 1553.3 meV Mahan exciton undergoes abrupt, fundamental changes: Its binding energy decreases to 1.5 meV, its oscillator strength decreases sharply, the linewidth of the 1553.3 meV exciton enhancement decreases from 2.7 to 0.9 meV, and a double-peak PL spectrum which is indicative of biexciton formation [29] is observed. These changes suggest the possibility that for $n_s \approx 1.9\times10^{11}$ cm^{-2} many-body interactions result in a large increase in the radiative lifetime of the 1553.3 meV excitons which enables them to condense and form biexcitons. At $n_s \approx 1.9\times10^{11}$ cm^{-2} the exciton ground-state energy and binding energy change discontinuously to 1550.2 and 4.1 meV, respectively. The fact that we simultaneously observe the 1553.3 and the 1550.2 meV excitons only over the narrow range -47 V $\leq V_g \leq -42$ V (corresponding to $n_s \approx 1.9\times10^{11}$ cm^{-2}) is significant and can be explained by a 1.5% variation in n_s across the excited area of our sample, taking into account the above depletion rate. The 1553.3 meV exciton is not observed for $n_s < 1.9\times10^{11}$ cm^{-2} showing that it is not an allowed exciton eigenstate for this range of n_s. Over the range 1.4×10^{11} cm^{-2} $\leq n_s <$ 1.9×10^{11} cm^{-2} the exciton ground-state energy remains unchanged at 1550.2 meV. Figure 2(b) shows that only free e-h PL is observed at $V_g = -147$ V. The unbinding of the Mahan exciton at $V_g = -147$ V might be due to the deformation of the SHQW wave function and the resultant increased scattering of the 2DEG. These results conclusively show that the Mahan exciton eigenstate energy is sensitive to n_s only at the critical density where it undergoes a sharp discontinuity. The physical mechanism for this discontinuity is not understood at present. Nevertheless, we believe it is a manifestation of the true many-body nature of the Mahan exciton eigenstate. A recent theory predicted that the bulk exciton energy is independent of electron density due to the cancellation of self-energy and vertex corrections [9]. Our results clearly show that other many-body effects have to be taken into account to explain the observed discontinuity in the energy of the two-dimensional Mahan exciton.

In summary, we report the first observation of Mahan excitons in the low-temperature photoluminescence and magnetophotoluminescence spectra of a degenerate quasi-two-dimensional electron gas. The spectra exhibit a sharp Fermi surface and a well-resolved exciton resonance which is sensitive to electron density and temperature. We observe a sharp decrease in the exciton linewidth and oscillator strength with a concomitant double peak spectrum which is attributed to the formation of biexcitons and a large discontinuity in the exciton ground-state energy at $n_s \approx 1.9\times10^{11}$ cm^{-2}. An abrupt transition from excitonic to free-electron-hole recombination occurs at $n_s \approx 2.2\times10^{11}$ cm^{-2}.

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