Observation of a Many-Body Edge Singularity in Quantum-Well Luminescence Spectra

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The observation of a many-body, Fermi-energy edge singularity in the low-temperature photoluminescence spectra of InGaAs-InP quantum wells is reported. Strong enhancement of the photoluminescence intensity towards the electron Fermi energy (E_F^e) is observed, due to multiple electron-hole scattering processes to states above E_F^e. Recombination of electrons in states up to E_F^e is allowed by hole localization. The many-body processes are analogous to the core-hole phenomena in the soft-x-ray emission spectra of metals.

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The observation of a strong enhancement in the luminescence intensity for electrons recombining close to the Fermi energy (E_F^e) in n-type, modulation-doped, InGaAs-InP quantum wells (QW's) is reported. This is a direct manifestation of the enhanced, multiple electron-hole (e-h) scattering rate for electrons close to E_F^e in an electron plasma of density $n_s^e \sim 10^{12}$ cm^{-2}. Such scattering processes for electrons at energy $E \ll E_F^e$ are suppressed by the exclusion principle, so that enhancement occurs only close to the sharp Fermi edge which exists at low temperature. Although electron-hole bound states (excitons) do not exist at high n_s^e because of screening and phase-space occupation, the experiments demonstrate that e-h Coulomb interactions are still very important, and play a dominant role in the determination of the low-temperature luminescence line shapes. The many-body Fermi-energy enhancement is analogous to that discussed for metals and degenerate semiconductors nearly twenty years ago by Mahan[1,2] and by Nozières and de Dominicis,[3] and observed previously only in the soft-x-ray emission and absorption spectra of metals such as Na,[4] Al, or Mg,[5] where it is referred to as the many-body "x-ray edge singularity."

In the x-ray-emission case an electron in the conduction-electron Fermi sea recombines with an inner-shell, "core" hole. k conservation in the recombination is ensured because the core hole is strongly localized in real space and has sufficient spread of k vector to enable electrons at E_F^e (with wave vector $k_F^e \sim 1\times10^8$ cm^{-1}) to recombine. In the present case the holes in the InGaAs-InP QW's are sufficiently localized, probably by alloy fluctuations, to permit all the electrons up to E_F^e to recombine without significant restriction due to k conservation. This contrasts with the situation in modulation-doped GaAs-GaAlAs QW's where the low-temperature photoluminescence (PL) spectra are dominated by electrons recombining close to $k_e = 0$, because of the limited range of the k vector of the low-density photocreated holes.[6] Fermi-energy enhancements have been predicted for the absorption of luminescence spectra of highly excited semiconductor QW's by Schmitt-Rink, Ell, and Haug,[7] and for modulation-doped QW's by Ruckenstein, Schmitt-Rink, and Miller.[8] For highly excited QW's, k conservation is ensured because equal numbers of high-density electrons and holes are photocreated. Experimentally it may be difficult to maintain sufficiently low sample temperatures, under the high-excitation conditions necessary,[9] to observe the many-body effects due to e-h multiple scattering.

The present experiments were carried out on $In_{1-x}Ga_xAs$-InP modulation-doped, single QW's grown close to lattice match ($x = 0.47$) by atmospheric pressure metal-organic chemical vapor deposition.[10] The structure consisted of a semi-insulating InP substrate, a 200-Å undoped InP buffer layer, 500 Å of InP doped at 5×10^{17} cm^{-3} with sulfur, a 100-Å undoped InP spacer layer ($n \sim 5\times10^{15}$ cm^{-3}), the InGaAs QW of 100-Å width ($n \sim 2\times10^{15}$ cm^{-3}), a 100-Å InP spacer layer, and finally 300 Å of doped InP ($n \sim 5\times10^{17}$ cm^{-3}). An electron density in the QW of 9.1×10^{11} cm^{-2} is found from low-temperature (2-K) Hall measurements under illumination.[11] PL was excited by direct excitation of the QW by $\sim$20 mW/cm^2 of the 1.09-μm line (1.14 eV) of an Ar-ion laser. On the assumption of a carrier lifetime of 1 nsec this corresponds to the creation of a minority-carrier (hole) density of $n_s^h \simeq 10^8$ cm^{-2}.

Typical PL spectra from 4.7 to 80 K are shown in Figs. 1(a)–1(d). The spectrum at 4.7 K is composed of a low-energy onset at $\sim$840 meV of width $\sim$10 meV, a slowly varying region over the next 10 meV, then a strong increase towards the high-energy cutoff at $\sim$885 meV. The overall width of the PL band of 44.1 meV [see Fig. 1(a)] is very close to E_F^e of 45.4 meV calculated from $n_s^e = 9.1\times10^{11}$ cm^{-2}, by use of an electron effective mass of $0.048m_0$.[12] We conclude that recombination is observed for electrons throughout the Fermi sea, with maximum PL intensity occurring for electrons at E_F^e with $k_F^e \sim 2.4\times10^6$ cm^{-1}. A very improbable, nonthermalized free-hole distribution with a temperature of order 200 K [13] would be required to give efficient recombination for electrons at E_F^e if only vertical, k-conserving transitions between free-particle states occurred. How-

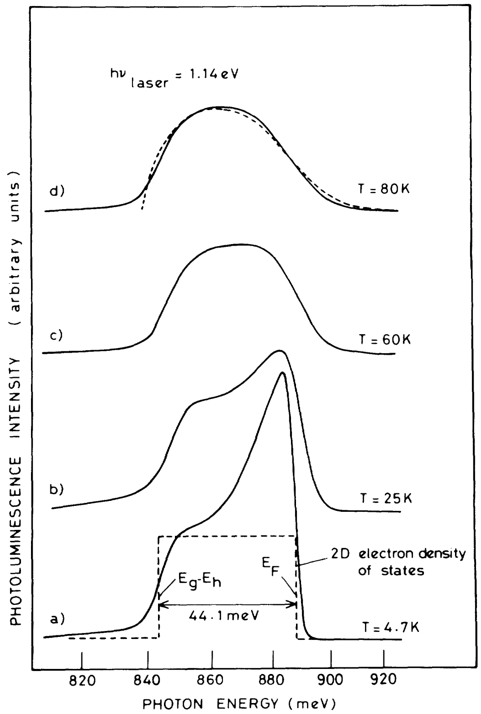

FIG. 1. PL spectra at 4.7, 25, 60, and 80 K from a modulation-doped InGaAs-InP quantum well with $n_s = 9.1 \times 10^{11}$ cm^{-2}. The spectrum at 4.7 K in (a) shows the strong enhancement towards the Fermi energy E_F. The theoretical spectrum from Eq. (1) corresponding to the ideal 2D density of states is superimposed to emphasize the Coulomb enhancement effects. With increasing temperature the enhancement effects are reduced, and are absent at 80 K where a good fit is obtained assuming no enhancement [the dashed curve in (d)].

ever, the abrupt Fermi cutoff in Fig. 1(a) (E_F on the figure) shows that the carrier temperatures must be low (~ 10 K), and so we deduce that the e-h recombination is dominated by transitions between the high-density electrons and low-density (localized) holes.

Further information on the nature of the states involved in the recombination is provided by the spectra in Fig. 2 taken at magnetic fields (H) of 0 and 3.1 T, at 4.2 K. At 3.1 T, the spectrum has broken up into a series of equally spaced Landau levels (LL's) with spacing given by the electron cyclotron energy $eH/m_e c$, with $m_e = 0.048 m_0$. The observation of equally spaced LL's (up to 10 T) shows that the electron states are not perturbed by disorder or localization effects so that the skewed line shape of Fig. 1(a) cannot be attributed to a modified electron density of states. In addition, it demonstrates that the PL spectrum is not due to the superposition of two bands of unrelated origin. The absence of any hole LL splittings (up to 10 T) shows that only discrete bound-hole levels are populated, and that any contribution to the line shape due to the population of the anomalous hole density of states away from $k_h = 0$ can be ex-

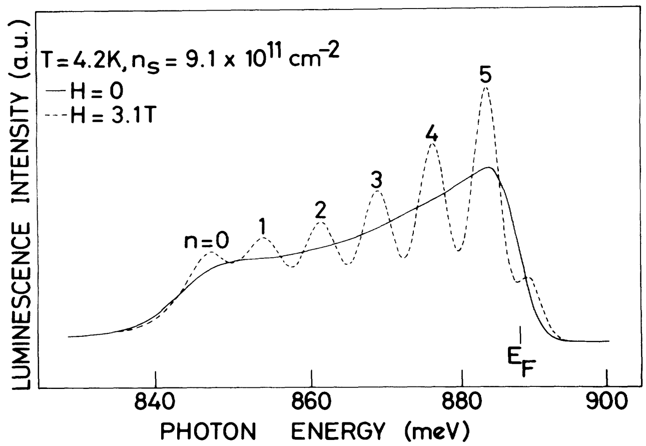

FIG. 2. Low-temperature PL spectra at $H = 0$ and 3.1 T. At $H = 3.1$ T, equally spaced electron Landau levels are observed, showing that unperturbed free electron levels are involved in the recombination, and that the spectrum does not arise from the superposition of two separate bands. The Fermi energy E_F at $H = 0$ is marked on the figure.

cluded.[13]

Strong evidence for hole localization in PL, of magnitude 12 ± 3 meV, at high n_s, in the same modulation-doped QW has been presented previously from a comparison of transition energies in PL and photoconductivity (equivalent to absorption) in magnetic field.[14] The localization probably arises because of alloy fluctuations of dimension ~ 50 to 200 Å, observed directly by transmission electron microscopy.[15] A radius of the localized hole wave function of 10 to 30 Å has been deduced from the magnitude of the strong exciton–LO-phonon coupling observed in the well.[15] With the use of the uncertainty relation $\Delta r \Delta k = \frac{1}{2}$, this range of radii corresponds to a range of hole wave vectors of $(1.6-5) \times 10^6$ cm^{-1}, of the order of the value required ($k_F^e = 2.4 \times 10^6$ cm^{-1}) to give efficient recombination, without k restriction, for electrons in all states up to E_F^e.

If we neglect many-body interactions, the PL line shape $[I_0(\omega)]$ is given by the convolution of the two-dimensional (2D) electron and hole densities of states (DOS) $[D_e(E_e), D_h(E_h)]$, multiplied by their respective Fermi functions $[f_e(E_e), f_h(E_h)]$, where

$$I_0(\omega) = \int \int D_e(E_e) f_e(E_e) D_h(E_h) f_h(E_h) g(E_e, E_h) \times \delta(\omega - E_e - E_h) dE_e dE_h. \quad (1)$$

E_e, E_h are the electron and hole energies above their respective band edges, ω is the energy from the onset of the PL band, and $g(E_e, E_h)$ is the oscillator strength without many-body interactions. For $T = 0$ the expected line shape is superimposed on Fig. 1(a) for $g(E_e, E_h) = 1$ (independent of E_e for an infinitely localized hole, radius $r_h = 0$) and corresponds to the steplike 2D electron DOS, with total width given by E_F^e, since $E_F^h \approx 0$. The main difference between the experimental line shape and $I_0(\omega)$ is the strong enhancement of the experimental spectrum towards E_F^e.

We now calculate the PL line shape including the effect of multiple *e-h* scattering and static screening within the Bethe-Salpeter equation in a simplified treatment of the 2D problem, taking $m_h=\infty$ (infinite hole localization). We include only ladder diagrams, neglecting the "crossed" diagrams which should be included for finite n_s. The effect of the crossed diagrams is to reduce the predicted logarithmic singularity at E_F to a power-law form. When the inhomogeneous broadening is of the same order as the binding energy of the exciton, as in the present case, the crossed diagrams do not have a major effect on the experimental line shape. The treatment results in a PL line shape of the following form:

$$I(\omega)=I_0(\omega)\{1-\Lambda_2(p)/[1+\Lambda_1(p)]\}^2, \qquad (2)$$

where the term in curly brackets is the excitonic enhancement factor, first developed by Mahan,[1] and Λ_1 and Λ_2 are defined in Refs. 1, 2, and 7. Full details of the theory will be presented elsewhere.[16]

In Fig. 3 the calculated PL line shape (the dashed line) from Eq. (2) at $T=4.7$ K is shown, including an inhomogeneous broadening of 5 meV due to well width and alloy fluctuations found in similar QW's at $n_s=0$,[14]

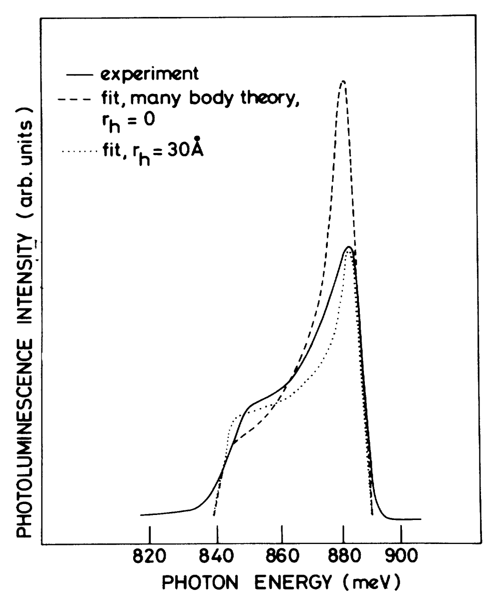

FIG. 3. Comparison of the low-temperature (4.7-K) PL spectrum of Fig. 1(a) with theoretical curves (with 5 meV inhomogeneous broadening) obtained from Eq. (2) for no k restriction for electrons up to E_F (very strongly localized hole $r_h=0$, the dashed curve), and $r_h=30$ Å (the dotted curve). The good agreement between theory and experiment provides strong evidence in favor of the many-body explanation of the PL line shapes.

and $g(E_e,E_h)=1$ as above. The other input parameters are $m_e=0.048m_0$, the low-frequency dielectric constant $\epsilon_0=13.8$, and the quasi-2D excitonic binding energy E_b of 6.2 meV. The correct form of the experimental spectrum, with the enhancement at E_F^e superimposed on the 2D DOS, is reproduced by the theory, although the calculated E_F^e enhancement is about a factor of 2 too large. If phenomenological account is taken of the finite r_h from calculation of $g(E_e,E_h)$ by our taking the Fourier transform into k space of the hole wave function in real space, the dotted curve in Fig. 3 is obtained for r_h taken equal to 30 Å. The effect is to reduce $I(\omega)$ at E_F by about a factor of 2 because of the limited range of hole wave vectors available. The remaining discrepancy in Fig. 3 is probably related to the neglect of crossed diagrams, final-state interactions such as intraband electron-hole pair formation and indirect transitions[8] for finite m_h, and the assumption of static screening and pure two dimensionality. Disorder effects will also broaden the electron momentum distribution, and lead to a reduction of the enhancement. In the light of these remarks, we believe that the qualitative agreement between theory and experiment in Fig. 3 provides convincing evidence for the role of *e-h* multiple scattering in determining the PL line shape.

With increasing temperature, the sharp Fermi level is broadened out, and the enhancement at E_F^e will be strongly reduced at temperatures of the order of $E_b/k_B\approx 70$ K as also predicted for highly excited QW systems with $n_e=n_h$.[7] We calculate only a 20% enhancement at E_F^e at 60 K, close to the experimental findings [Figs. 1(b)–1(d)], where the enhancement is no longer visible above ~70 K, providing further strong support for the model. Indeed, good agreement between the experimental spectrum and the $I_0(\omega)$ term at 80 K [Eq. (2) with no enhancement] is obtained, as shown by the full and dashed lines in Fig. 1(d).

The present work is the first convincing observation of the many-body "edge singularity" in semiconductor luminescence spectra. There are a number of reasons why the present system is particularly suitable for the observation of this effect. First, the occurrence of hole localization, probably by alloy fluctuations, permits recombination of all electrons up to E_F^e, in an analogous way to the core-hole problem in metals. It also removes any broadening of the high-energy cutoff due to hole recoil effects.[8] Second, the modulation doping of the QW structure, unachievable in the 3D case, strongly reduces scattering effects by the dopants. Third, the *e-h* Coulomb interaction is enhanced and its screening at high n_s is reduced, in 2D relative to 3D, thus increasing the magnitude of the phenomenon in 2D. PL from modulation-doped InGaAs wells (clad by AlInAs) has been reported by Penna *et al.*[17] They observed a PL spectrum increasing in intensity towards E_F, but the spectra were too broadened by inhomogeneous processes to draw any conclusions about *e-h* interactions. PL from the 2D electron gas in a Si inversion layer recombining with holes on neutral acceptors in the bulk of the material has been studied by Kukushkin and Timofeev.[18] In this case, the 2D DOS was reproduced in the PL spectrum, but without the enhancement at E_F^e probably because the *e-h* overlap is much weaker since the electrons and holes are spatially separated.[19]

To conclude, the observation of a Fermi-energy edge singularity in the luminescence spectra of modulation-doped InGaAs-InP quantum wells has been reported. Good qualitative agreement is found between the spectral line shapes and many-body calculations. The phenomena are closely related to the excitonic enhancements at E_F found in x-ray emission and absorption spectra of metals.

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