Doping-density dependence of photoluminescence in highly Si-doped $GaAs/Al_xGa_{1-x}As$ quantum wells from below to above the metallic limit

C.I. Harris and B. Monemar

Max-Planck-Institut für Festkörperforschung, Heisenbergstrasse 1, 7000 Stuttgart 80, Federal Republic of Germany and Department of Physics and Measurement Technology, University of Linköping, Linköping S-58183, Sweden

H. Kalt

Max-Planck-Institut für Festkörperforschung, Heisenbergstrasse 1, 7000 Stuttgart 80, Federal Republic of Germany and Fachbereich Physik, University of Kaiserslautern, Kaiserslautern, Federal Republic of Germany

K. Köhler

Fraunhofer Institut für Angewandte Festkörperphysik, Eckerstrasse 4, Freiburg, Federal Republic of Germany (Received 29 December 1992; revised manuscript received 24 March 1993)

The development of the photoluminescence spectra with doping density has been studied for a series of single $GaAs/Al_xGa_{1-x}As$ quantum wells center doped with Si. The trends observed are found to be very different from those observed for equivalent doping levels in bulk GaAs. In particular, excitons continue to dominate the radiative recombination at doping levels right up to the metallic limit. The electron density at which the exciton is quenched is found to be considerably higher (about $10^{12}~\rm cm^{-2})$ in the center-doped case than has previously been demonstrated for modulation doped quantum wells (about 4×10^{11} cm⁻²). This result is understood in terms of the difference in phase-space filling for the center-doped quantum well when compared with the modulation-doped case. The impurity concentration at which the material becomes degenerate is also found to be significantly higher than in bulk GaAs. The recombination dynamics at the metallic limit have been studied using a time-resolved photoluminescence technique and demonstrate that localization, resulting from either interface roughness or the high doping level, is reduced as the density is increased above the degenerate limit. The apparent relaxation of momentum conservation observed in the photoluminescence spectra for highly doped samples is interpreted as being due to recombination from strongly localized holes.

I. INTRODUCTION

The properties of semiconductors, both optical and transport related, vary substantially as the level of doping is increased. For high dopant concentrations a regime is reached in which the material is said to be "degenerate." This threshold corresponds to the onset of overlap of the broadening impurity band with the free-carrier continuum, in turn, representing a transition from insulating to metallic behavior. The metal-insulator transition has been studied optically in bulk GaAs for some 30 years.^{1-8} A detailed understanding of the electronic phase change taking place at high doping concentrations in GaAs has not yet, however, become clear. At doping levels well below the metallic limit in bulk GaAs [threedimensional (3D)] exciton related transitions no longer appear in the optical spectra at low temperature, even under low-intensity excitation conditions; this effect has been associated with the strong screening of the electronhole $(e-h)$ Coulomb interaction by carriers and impurities. The typical concentration of impurities in bulk GaAs necessary to completely suppress the exciton (free or bound) at low temperature is found to be around 10^{16}

 $\text{cm}^{-3.9}$ Optical spectra for higher doping levels are then dominantly band-to-band and band-to-impurity related transitions.

The doping level needed to reach the metallic transition has not been accurately determined for bulk GaAs. The difficulty in making a definitive measurement of dopant concentrations, 10 particularly at high doping levels where compensation effects question the use of methods based on the number of ionized carriers, seems to have excluded to date a systematic study correlated with adequate transport measurements. It is apparent from a survey of optical spectra for n-type GaAs obtained by different authors that the transition occurs at a doping concentration around 5×10^{17} cm⁻³, i.e., a little higher than the theoretical limit calculated from the onset of wavefunction overlap ($\sim 2 \times 10^{17} \text{ cm}^{-3}$).^{2,3,5,7,8,11} Above this limit, optical spectra are dominated, for n-type GaAs, by transitions from the degenerate electron population in the conduction-band-to-acceptor or valence-band states. One issue of continuing debate is whether it is necessary to relax the requirement for momentum conservation in order to account for the observed photoluminescence (PL) line shape.^{7,8,12} In an *n*-type quantum well the in-

0163-1829/93/48(7)/4687(8)/\$06.00 48 4687 61993 The American Physical Society

elusion of nonmomentum conserving recombination will result in a well-defined line shape reflecting the steplike density of electron states. The observed PL peak position is determined by a combination of two major effects, the shift upwards in photon energy due to band filling up to the Fermi level ("Burstein-Moss" shift) and the counteracting effect of reduction in band gap due to many-body effects of the interacting carriers (the so-called band-gap renormalization). These shifts are of comparable magnitude, but for n-type GaAs the Fermi-level shift dominates, leading to an upward shift of the PL peak with increased doping above the metallic limit, $2,3,5,7,8,11$ while in p-type GaAs the reverse situation is true.^{2-4,6,8}

In the two-dimensional (2D) case, specifically for $GaAs/Al_xGa_{1-x}As quantum wells (QW's), there has$ been little discussion in the literature of the situation where the doping inside the well is increased up to the metallic limit. A major difference between the recombination in 2D and 3D systems is that the excitons are significantly stronger in the optical spectra of the 2D case. The screening of 3D excitons, which is already strong at very low doping and carrier densities, is much weaker in the 2D case. Work on modulation-doped 2D structures has shown that the lowest-energy excitons can exist simultaneously with free carriers up to sheet carrier densities of about 4×10^{11} cm⁻².¹³⁻¹⁵ At higher carrier densities, the excitons are quenched due to a combination of many-body-related effects, carrier screening due to Coulombic interaction, short-range exchange and correlation, $16,17$ and phase-space filling by free carriers. Higher-energy exciton states can still be clearly observed in absorption, demonstrating that exciton screening is relatively inefficient.¹⁸⁻²¹ The effect of center doping in the quantum well on the screening and correlation of carriers has received considerably less attention. At low doping the presence of excitons both free and bound, and their dependence on the well width and the position of the dopant, have been extensively studied.²² The properties of highly doped quantum wells ($\sim 10^{12}$ cm⁻²) have previously been discussed in the case of an $c²$) have previously been discussed in the case of an interesting anomalous polarization of the spectra found $for such structures.²³ This behavior has been explained$ in terms of many-body interactions within the electron Fermi sea. 17 No systematic study of the influence of the doping level in the well on the optical spectra of a QW structure, however, seems to have been presented so far in the literature.

In this paper we discuss such a systematic study of the optical properties of *n*-type Si-doped 100- \AA QW's, where the doping density has been varied between 2.5×10^8 and 2.5×10^{12} cm⁻² (3D density $\sim 5 \times 10^{14} \rightarrow 5 \times 10^{18}$). The study concentrates on the high doping region, i.e., on the transition from exciton to free-carrier behavior in the optical spectra. Steady-state PL and PL excitation (PLE) data are complemented with picosecond transient studies performed with a streak camera unit, in order to reveal the dynamics of the exciton and free-carrier systems in the high doping regime.

A dopant placed in a layered structure such as a QW has the possibility, given the appropriate band structure, of losing its charge to an adjacent layer; this is the wellknown principle of modulation doping. For a centerdoped structure this effect is generally considered to be negligible; however, charge loss can occur given the presence of states of lower energy in the adjacent layers. For an n-type doped system such states would be deep levels or shallow acceptors. This charge loss will in general have a small effect on the band structure of the system. If, however, there exists a high density of "accepting" states the dopants can be entirely depleted resulting in a strong Coulombic potential due to the ionized. centers. One example of this has been studied by Harris ${\it et}~al.^2$ in which depletion of a doped quantum well results from a high density of defect states at the nearby surface. In this work we also illustrate how the depletion of charge from the quantum well results in a strong distortion of the confining potential at high doping levels.

The paper is organized in the following way. Section II briefly describes the experimental technique and the sample structure investigated. Section III presents the experimental data, including both steady-state PL and PLE data and picosecond time-resolved PL spectra. The development of the optical spectra with doping density is discussed, in particular concentrating on the behavior just at the metallic limit. A study of samples above the degenerate limit is also presented. In Sec. IV we discuss the results, making a direct comparison with previous data from the 3D case (highly doped GaAs) to illustrate the main differences. Finally, Sec. V makes some summary conclusions from this work.

II. SAMPLES AND EXPERIMENTAL PROCEDURE

The series of samples used for this study were grown by molecular-beam epitaxy. The structure is a single quan- ${\rm tum}$ well of 100 Å well width, the barriers are 150 Å wide with a 34% Al content. A GaAs buffer layer and a subsequent short-period superlattice have been grown before the QW structure to minimize background impurity levels. All layers except the active QW were nominally undoped. The active 100-A. QW layer was doped in the central 50 A by continuous addition of silicon during growth and without growth interruption at the interfaces. The growth temperature was chosen to be 680° C. This is a compromise between high temperatures, which are known to producer higher interface quality, and low temperatures used to minimize segregation effects, which spread the doping profile within the QW in the direction of growth. At this growth temperature some interface defects are introduced during growth, leading to a nonradiative recombination channel. A bulk doping level within the 50-Å doped region of from 5×10^{14} cm⁻³ to 5×10^{18} cm⁻³ is covered, corresponding to sheet densities of 2.5×10^8 cm⁻² to 2.5×10^{12} cm⁻² in the QW.

The time-integrated photoluminescence data were obtained at 1.6 K with a photon-counting detection technique, employing a Spex double monochromator. PLE spectra were obtained with tunable dye laser excitation using an LD700 dye pumped with a Kr^+ ion laser. The picosecond time-resolved data were obtained with a synchroscan streak camera in combination with a 0.32-m spectrometer; the time resolution for the combined system is of the order of 10 ps.

III. EXPERIMENTAL RESULTS

A. Optical spectra as a function of doping level

The strong dependence of the low-temperature PL spectra upon doping concentration in the QW is illustrated in Figs. 1 and 2. The results from samples with $\rm{doping~in~the~range}~up~to~3\!\times\!10^{18}~cm^{-3}~(1.5\!\times\!10^{12}~cm^{-2})$ are shown. In both cases the substrate luminescence has been subtracted for clarity. As discussed in the Introduction, two sets of data are reported. Figure 1 illustrates spectra taken with excitation at 5145 \AA ; under these conditions the quantum well is believed to have essentially fiat-band conditions, i.e., the Si donor is not ionized. The generation of electron-hole pairs with energy larger than the $\text{Al}_x\text{Ga}_{1-x}$ As barrier band gap allows a redistribution of charge to compensate for the internal field brought about by charge transfer between impurities in different layers.²⁴ Excitation at energies below the barrier does not allow for the transfer of charge and depleted conditions obtained at higher temperatures and preserved during cool down of the sample are maintained (Fig. 2).

Figure 1, then, illustrates the development in PL spectra for increasing doping density with the donor population in a mostly neutral charge state. The neutral donor bound exciton is not resolved for any of the samples shown; its presence is indicated by the asymmetric broadening to lower energies of the free exciton peak (the broadening to lower energies is most clearly seen at a doping density of 2×10^{16} cm⁻³). The linewidth of the exciton transition is found to increase with doping den-

FIG. 1. Dependence of optical spectra on doping density (5145-A excitation). Approximate normalization terms are indicated.

FIG. 2. Dependence of optical spectra on doping density (7400-A excitation). Approximate normalization terms are indicated.

sity. In addition, a broad donor-to-valence-band transition appears extending from 1.545 eV down to a weak low-energy tail, as low as 1.45 eV for the most highly doped samples.

The broadening at the low-energy tail of the donor-tovalence-band transition can be associated with the strong localization of the holes; such localization is a consequence of the random potential distribution produced by a high doping density. Compensation, which enhances the potential fiuctuations in a material due to the ionization of acceptors and donors, will play an important role in increasing the localization in these samples. The Si dopant used is amphoteric and at high densities an increasing fraction occupies an acceptor (As) site. Between the doping levels of around 7×10^{17} cm⁻³ and 1×10^{18} cm^{-3} there is a distinct change in the PL spectra, which demonstrates an apparent reversal in the trend of broadening spectra and seems more consistent with a drop in the doping density. Interpreting this effect within the model of localization in a random potential requires an effective decrease in the size of the potential Huctuations. This is, however, inconsistent with the traditional picture of increased localization with doping density; indeed one would expect the strongest localization at the highest doping density where the additional process of compensation begins to be effective. We instead suggest that the spectra can be understood if we assume that this sample is already degenerate, but only marginally above the metallic limit. The presence of a mobile charge smears out the fluctuations in the potential due to the random distribution of donors, in turn reducing the localization. This interpretation of the experimental data also implies that the exciton still remains in the optical spectra even at the degenerate limit. In addition the picture is par-

ticularly attractive in that it implies a relatively abrupt transition to the metallic state, as is indeed observed. Such a mechanism has not been discussed for bulk material, although there a potentially stronger effect than for the 2D case would be expected, but would be observed less easily due to the absence of excitons in the recombination spectra.

The results of experiments with modulation-doped structures have demonstrated that the $n = 1$ exciton is quenched at free-electron concentrations greater than 4×10^{11} cm⁻². The degenerate limit proposed from the experimental data in this work would provide an equivalent sheet carrier concentration of 10^{12} cm⁻², significantly higher than the limit for the quenching of the $n = 1$ exciton demonstrated in the modulation-doped case. The mechanisms responsible for the loss of the exciton are therefore less effective in the center-doped case. The principal mechanisms behind the quenching of the exciton in the case of filling in a single subband are understood to be screening and phase-space filling, which result in a progressive unbinding of the exciton state¹³ (note that screening alone results in a finite binding energy in 2D). For center doping there exists an array of impurities which act as strong scattering sites and thereby reduce the effectiveness of screening. In addition the effect of a high doping density is to distort the shape of the density of states close to the band edge. As a result the corresponding occupancy of k space can be very different from that for the modulation-doped structure with the result that the exciton states are not blocked at the same limit. The uppermost spectrum in Fig. 1 clearly demonstrates the shifted emission edge at high energies characteristic of the Moss-Burstein effect due to band filling. At this high electron density 2.5×10^{12} cm⁻² the free-exciton transition is finally lost in PL and recombination occurs via band-to-band transitions.

 $\underset{\ast}{\text{Excitation}}$ at energies below the $\text{Al}_x\text{Ga}_{1-x}$ As band gap is shown in Fig 2. As previously discussed this set of data corresponds to the situation where a large percentage of the donors are ionized and there exists a strong internal electric field across the well. Two further consequences of the increased doping are apparent. First, the internal field becomes very strong at high doping levels; this contributes strongly to the quenching of the exciton luminescence. Second, at high doping levels there is a strong localization effect; this is due to the random distribution of ionized donor potentials and the increasing localization of holes at the QW interface with increasing field. The spectra at high doping density ($> 2 \times 10^{17}$ cm^{-3}) again show an apparent anomaly in the picture of strong localization. In fact, the data provide strong support for the model discussed so far regarding reduced localization above the degenerate limit. As implied, the sample with a doping concentration of 7×10^{17} cm⁻³ represents the strongest localization case, where the random potentials are not smeared out by free carriers. The two higher doped samples demonstrate progressively weaker localization as the number of free carriers in the well is increased. In both cases, however, there is no band filling present and the loss in exciton luminescence is associated with the combination of a strong internal field and localization. For the low doped samples there are clearly two peaks resolved; these probably result from the free exciton and either an enhanced donor-to-valence-band transition or an exciton bound to an ionized donor.²⁴

Figure 3 shows the corresponding PLE data for the conditions of Fig. 1. The spectra are measured with additional low-intensity excitation at high photon energy to achieve band conditions similar to those under which PL was measured. The retention of exciton character in the observed transition up to the highest doping level is again illustrated in the PLE data. In fact there is relatively little change in the spectra with doping level other than a slight decrease in the exciton contribution in comparison to the free-carrier level. The Stokes shift between PL and the heavy-hole exciton in PLE is found to increase from less than 1 meV at low doping to approximately 3 meV at the highest observable level. This is as expected from the increased localization with doping so far discussed. The data for the highest doped sample do not show any $n = 1$ exciton and are upshifted in energy due to band filling. The low-energy absorption tail corresponding to the position of the electron Fermi level is broadened to a greater extent than that expected from simple thermal broadening. The major contribution to this width results from impurity scattering and to a lesser extent hole localization at the valence-band edge (see following discussion).

B. Recombination dynamics at the metallic limit

The sample with a donor concentration in the well of $1.5\times 10^{12} {\rm cm^{-2}}~(\sim 3\times 10^{18}~{\rm cm^{-3}})$ represents the limiting case at which the lowest-energy $n = 1$ excitons are not observed, due to electron filling in the lowest conduction subband. As shown in Figs. 1 and 3 neither steady-state PL nor PLE spectra show any sign of the lowest exciton states; rather the PLE spectrum demonstrates a broad-

FIG. 3. Development in PLE with doping concentration. Additional low-intensity excitation at 5145 A provides comparable band conditions to Fig. 1.

ened steplike onset characteristic of a Fermi level above the band edge. The difference between the PL spectra for highly doped samples which still exhibit an exciton transition and those for which it is lost can be studied further using time-resolved PL. Figure 4 compares the short-time development in the PL spectra for two samples: (a) one slightly below the doping limit for which excitons are lost in PLE and (b) the second slightly above. In both cases

FIG. 4. Comparison of time-resolved PL spectra for samples at (a) and above (b) the metallic limit.

excitation is at a photon energy corresponding to the $n =$ 2 exciton and thus gives band-bending conditions across the structure similar to those described by the cw data in Fig. 2. Figure 4(a) illustrates that, although the exciton is not observed in the cw spectra, it is still seen for short times $({\sim 150 \text{ ps}})$ in the time-resolved measurement (FE/BE). This is consistent with the PLE data, which continue to show the exciton at this doping level. We believe that the rapid loss in the exciton component is brought about via the strong localization of the hole and the subsequent breakup of the exciton in the internal electric field. The continuing relaxation to lower energies of the PL spectra following the loss of the exciton illustrates that further localization is still taking place. The data for the sample above the exciton limit [Fig. 4(b)] have, in contrast, no short-lived component, but show a relatively weak but continuous localization. The slower relaxation is again indicative of the weaker-localization potentials present in the strongly degenerate sample. The effective lifetime for recombination in the two cases is also seen to be significantly different. Although the exciton component in Fig. 4(a) decays extremely rapidly, the remaining donor-to-valence-band transition is as expected relatively long lived ($\tau_{\text{eff}} \sim 1.56$ ns; note that this analysis is made difficult by the strong localization, manifested as a shift to lower energies with time of the PL, as a result we quote an intermediate value). In the strongly degenerate sample the decay of the band-to-band recombination $(B-B)$ is significantly faster (620 ps). This difference we again associate with the weaker localization, given that recombination can be expected to be more efficient in a system where higher mobility allows stronger carrier interaction. In addition the higher doped sample is found to show a proportionately larger nonradiative recombination term as indicated by temperature-dependent data. Given the complexity of the recombination mechanisms involved we avoid the discussion here of the dynamics in terms of a quantitative radiative lifetime for which it is extremely difficult to derive a meaningful value. Even in the lower doped case the evaluation of true lifetime parameters requires the solution of a system of coupled differential equations which include terms describing the many interaction mechanisms. An in-depth discussion of this kind of analysis is given in Refs. 25 and 26.

To the authors' knowledge the corresponding dynamics have not yet been properly studied for highly doped bulk GaAs. It is clear, however, that the recombination scenario in this material would be fundamentally diferent at the same limit. As pointed out, excitons are already lost at densities well below the metallic limit; for this reason there is no equivalent transition from exciton to non-exciton-type recombination occurring in the same density regime as the impurity Mott transition. It is of interest, however, to consider the role of localization and screening in the dynamics at this limit. The current work suggests that a significant decrease in the low-temperature radiative lifetime would be expected in bulk material for doping densities above degeneracy as compared to doping levels just below this limit. This result would be independent of any dependency of a nonradiative channel on doping level. We hope to address

these questions in the near future. A system that can be usefully compared is the equivalent quantum structure, but with modulation doping; here there is no significant impurity concentration in the well and a high density of a single charge type only exists. As already discussed this results in a different limit at which true excitons are lost. Localization, which dominates the recombination dynamics in the center-doped case, is not effective at high densities in modulation-doped structures. At short times $(<100 \text{ ps})$ following pulsed excitation additional band filling can be observed in the time-resoved PL spectra in this modulation-doped case. There is, however, no equivalent short-lived exciton component observed in samples for which the exciton is no longer observed in the cw spectra. 27 Lifetimes in such structures are typically a few hundred picoseconds, i.e., comparable to the center-doped structure at the highest density.

IV. DISCUSSION

Comparison of 2D and 3D behavior at high doping levels

In discussing the results obtained it is useful to compare the properties of highly doped QW's with the corresponding properties of bulk semiconductors in the same doping range. One considerable difference obvious from the results of this work is a higher metallic limit in the $\rm QW$ case, a concentration of about $2.0\!\times\!10^{18}~\rm cm^{-3}$ (corre- $\rm{sponding\ to\ a\ sheet\ density\ of\ about\ 1\times 10^{12}\ cm^{-2}).\ This}$ is a factor ²—3 times higher than in the 3D case. A simple interpretation for this higher limit in the QW is the considerable difference in donor binding energy between the QW and bulk GaAs. While the shallow donor binding energy in GaAs is about $5.9 \text{ meV},^{28}$ it is increased by a little over a factor of 2 to about 12 meV in a 100-Å QW, due to the confinement of the donor wave function.²⁹ There is no evidence to suggest that "self-screening" takes place at higher doping levels, which would reduce the donor $\rm binding$ energy. $\rm ^{30,31}$ This higher binding energy means that the donor impurity band created at higher doping levels can be broader in energy in the QW and sustain more donor electrons until the distribution merges into filling the conduction band in the metallic limit.

Another dramatic difference between bulk and 2D obvious from this work is the behavior of excitons vs doping level. In bulk GaAs excitons are already screened at a donor doping level of a few 10^{16} cm⁻³. Above this doping level no excitons are observed, either in PL emission or in absorption. This is very different from the QW case, as already pointed out above. Even at doping levels well above the metallic limit, i.e., $2.5 \times 10^{12} \text{ cm}^{-2}$ $(5 \times 10^{18} \text{ m})$ cm^{-3}), excitons are still found to exist.³² Similarly for modulation-doped structures, although the $n = 1$ exciton is lost at carrier densities of around 4×10^{11} cm⁻², the $n = 2$ exciton is still observed at densities as high as 7.5×10^{11} cm^{-2.33} It is clear that the way in which the excitons are screened is fundamentally difFerent in the confined system in comparison to bulk material.¹³ The argument for the lower screening strength in the low-dimensional system follows from the reduced degree of movement available to the carrier. Since screening is

basically a many-body effect where the carriers arrange themselves to maximize the total Coulombic interaction and hence minimize the single-particle interaction, any restriction on movement will inhibit their ability to do this. The same qualitative reasoning explains the limited screening due to impurity bound charge. The ineffectiveness of screening in 2D has been discussed on a number of occasions; $34-36$ it is clear that the weakness of this mechanism is the main reason for the retention of exciton recombination at high doping levels in the 2D system.

The reason for the higher carrier density at which the exciton is quenched for the center-doped case when compared to the modulation-doped structure is, however, less clear. The difference can be understood if one considers the occupation of phase space in the two cases. From efFective-mass theory the bound donor states are made up from contributions from states in the conduction band. The free carriers introduced by modulation doping occupy states from the band minimum upwards, whereas donor bound electrons contribute to blocking of states over a distributed range in k space, this distribution being density dependent. As a result the limit of phase-space filling for which the states that go to form the exciton are blocked can be significantly larger in the center-doped case. Furthermore, the differences in binding energy for donors and acceptors implies that the limit for acceptors will be larger than that for donors, a point currently under investigation. The effect of impurity density and binding energy on the k-state distribution, the so-called spectral density, has been discussed for GaAs quantum wells by Gold, Ghazali, and Serre.³⁷ Their calculations illustrate that there is a substantial spread in k space extending to higher energies from the band edge. As a rough approximation one can calculate this extension in k given that the spectral density is significant for $k < 1/a^*$ (in fact, from the calculations of Gold, Ghazali, and Serre $k < 0.6/a^*$ for a 100-Å well). Assuming a free-electron mass of $0.0667m_e$, this gives an extension in energy of approximately 5.6 meV above the band edge. An alternative but equivalent view (in real space) of the effect of shallow impurities on phase-space filling is to describe the distortion in the density of states resulting from increasing doping levels.^{38,39} At high densities the formation of band tails in the forbidden gap limit the extension of the Fermi level above the defined band edge and hence the exciton remains to higher densities.

The mechanisms of band-to-band recombination, as observed for the highly doped sample, are also best discussed. in comparison with bulk mechanisms. A point of continuing debate for highly doped GaAs is whether the explanation of the observed photoluminescence line shape requires the absence of momentum conservation in the radiative transitions or not.^{7,8,12} It has been argued that strong impurity scattering can lead to an enhancement in nonmomentum conserving transitions.⁴⁰ For the modulation-doped case work to date has clearly demonstrated that momentum conservation is fully obeyed.⁴¹ In the present system, however, the picture is again unclear since there exists the same strong scattering potentials as exist for the bulk. Figure 5 illustrates the pho-

FIG. 5. Recombination for highly doped quantum well. The broad plateaulike shape corresponds to the 2D density of states.

toluminescence spectra of a highly degenerate sample. A broad well-defined fIat region is observed extending over a large energy range. Such a plateau over a much larger photon energy range has been observed in similar samples with considerably higher dopant densities $({\sim 5\times 10^{12}~\mathrm{cm}^{-2}}).$ The plateau in the emission spectra reflects the two-dimensional density of occupied electron states and implies recombination of electrons with wave vector $k > 0$. This can occur via either of two mechanisms: (i) a breakdown in momentum conservation in the radiative recombination due to strong impurity scattering or (ii) due to the localization of the hole there is sufficient spread in k space to allow momentum conservation. The experimental results for the highly doped samples show significant tailing to lower energies, which is a clear indication that strong hole localization is indeed present. A similar mechanism has been observed in a modulation-doped system where an additional layer of acceptors is placed in the well thereby localizing the holes. 42

V. CONCLUSIONS

The development of low-temperature photoluminescence spectra with increasing doping level in a single quantum well is found to be qualitatively very different from the behavior in bulk material. The exciton dominates the QW luminescence right up to the degenerate doping limit and is not screened by Coulombic interaction with dopants as is the case for bulk material. This doping level also exceeds the carrier density limit for excitons measured for modulation-doped quantum wells. The filling of phase space, which is responsible for the loss of the exciton in the modulation-doped case, is less effective in the center-doped case. The bound nature of the donor electron implies that the averaged wave function for the electron state is distributed in k space, with the result that a sufficient proportion of states that go to form the exciton are not blocked. The metallic limit is reached only at dopant concentrations a factor of approximately 2 to 3 times higher than for bulk GaAs. A simple interpretation of this is the corresponding difference in binding energy of the donor for the confined system. We propose that even at the degenerate limit excitons are still present in the optical spectra of the QW. The degenerate limit is indicated by a clear reduction in localization above a certain doping concentration due to the presence of free carriers which smear out the localization potentials. Significantly above the metallic limit where the exciton is no longer observed in PL, the spectrum is made up of free-carrier band-to-band and donor-band-to-valenceband recombination. PL spectra at the highest doping level show a broad Bat emission indicating recombination from throughout the 2D density of occupied states. Momentum conservation considerations thus imply that either holes are sufficiently localized to provide the required spread in k space or that the strong impurity scattering results in a breakdown in momentum conservation. To date there has been relatively little work on high doping levels in low-dimensional structures, the current work has highlighted a number of areas which require further investigation. In particular a theoretical treatment of the mechanisms suggested in this paper is required.

- ¹N.F. Mott, *Metal-Insulator Transitions* (Taylor and Francis, London, 1974).
- 2° D.A. Cusano, Solid State Commun. 2, 353 (1964).
- 3 D.E. Hill, Phys. Rev. 133, 866 (1964).
- ⁴ J.I. Pankove, J. Appl. Phys. **39**, 5368 (1968).
- ⁵H.C. Casey and F. Stern, J. Appl. Phys. 47, 631 (1976).
- 6 D. Olego and M. Cardona, Phys. Rev. B 22, 886 (1980).
- ⁷Jiang De-Sheng, Y. Makita, K. Ploog, and H.J. Queisser, J. Appl. Phys. 53, 999 (1982).
- ⁸G. Borghs, K. Bhattacharyya, K. Deneffe, P. Van Mieghem, and R. Mertens, J. Appl. Phys. 66, 4381 (1989).
- $9J.$ Shah, R.F. Leheny, and W. Wiegmann, Phys. Rev. B 16, 1577 (1977).
- 10 G.E. Stillman and C.M. Wolfe, Thin Solid Films 31, 69 (1976).
- $^{11}{\rm T}.$ Lideikis and G. Treideris, Semicond. Sci. Technol. 4, 938 (1989).
- ¹²B.E. Sernelius, Phys. Rev. B 34, 5610 (1986); 34, 8696 (1986).
- ¹³S. Schmitt-Rink, D.S. Chemla, and D.A.B. Miller, Adv. Phys. 88, 89 (1989).
- 14 D.A. Kleinman, Phys. Rev. B 32, 3766 (1985).
- ¹⁵ A.E. Ruckenstein and S. Schmitt-Rink, Phys. Rev. B 35, 7551 (1987).
- ¹⁶A. Pinczuk, S. Schmitt-Rink, G. Daman, J.P. Valladares, L.N. Pfeiffer, and K.W. West, Phys. Rev. Lett. 63, 1633 (1989).
- ¹⁷ A.E. Ruckenstein, S. Schmitt-Rink, and R.C. Miller, Phys. Rev. Lett. 56, 504 (1985).
- ¹⁸H. Yoshimura, G.E.W. Bauer, and H. Sakaki, Phys. Rev. B 88, 10 791 (1988).
- ¹⁹D. Huang, H.Y. Chu, Y.C. Chang, R. Houdré, and H. Morkoc, Phys. Rev. B 88, 1246 (1988).
- ²⁰G. Tränkle, E. Lach, M. Walther, A. Forchel, and G. Weimann, Surf. Sci. 196, 584 (1988).
- 21 D. Huang, J.I. Chyi, and H. Morkoc, Phys. Rev. B 42, 5147 (1990).
- $22R$. Stepniewski, S. Huant, G. Martinez, and B. Etienne,
- $23R.C.$ Miller and A.C. Gossard, Phys. Rev. B 28, 3645 (1983).
- 24 C.I. Harris, B. Monemar, G. Brunthaler, H. Kalt, and K. Köhler, Phys. Rev. B 45, 4227 (1992).
- ²⁵C.I. Harris, H. Kalt, B. Monemar, P.O. Holtz, J.P. Bergman, M. Sundaram, J.L. Merz, and A.C. Gossard, Mater. Sci. Forum 8\$-87, 1363 (1992).
- ²⁶C.I. Harris, B. Monemar, H. Kalt, P.O. Holtz, M. Sundaram, J.L. Merz, and A.C. Gossard (unpublished).

- ²⁸C.J. Armistead, P. Knowles, S.P. Najda, and R.A. Stradling, J. Phys. C 17, 6415 (1984).
- W.T. Masselink, Y.-C. Chang, H. Morkoc, D.C. Reynolds, C.W. Litton, K.K. Bajaj, and P.W. Yu, Solid State Electon. 29, 205 (1986)
- ³⁰ J.A. Brum, G. Bastard, and C. Guillemot, Phys. Rev. B \$0, 905 (1984).
- 31 E. Hanamura, J. Phys. Soc. Jpn. 28, 120 (1970).
- ³²C.I. Harris, H. Kalt, B. Monemar, and K. Köhler, Surf. Sci. 26\$, 462 (1992).
- 33 The nondominant contribution of screening to the quench-

ing of excitons in modulation-doped structures is discussed in the following articles: R. Stepniewski, W. Knap, A. Raymond, G. Martinez, J.C. Maan, and B. Etienne, Surf. Sci. 229, 519 (1990); M. Potemski, R. Stepniewski, J.C. Maan, G. Martinez, P. Wyder, and B. Etienne, Phys. Rev. Lett. 66, 2239 (1991); D. Huang, H.Y. Chu, Y.C. Chang, R. Houdré, and H. Morkoc, Phys. Rev. B 38, 1246 (1988).

- ³⁴G. Tränkle, H. Leier, A. Forchel, H. Haug, C. Ell, and G. Weimann, Phys. Rev. Lett. 58, 419 (1987).
- ³⁵S. DasSarma, R. Jalabert, and S.-R. Eric Yang, Phys. Rev. B 41, 8288 (1990).
- ³⁶C. Delalande, G. Bastard, J. Orgonasi, J.A. Brum, H.W. Liu, M. Voos, G. Weimann, and W. Schlapp, Phys. Rev. Lett. 59, 2690 (1987)
- 37 A. Gold, A. Ghazali, and J. Serre, Phys. Rev. B 40, 5806 (1989).
- 38 Ulf Ekenberg, Phys. Rev. B 30, 3367 (1984).
- 39 B.I. Halperin and Melvin Lax, Phys. Rev. 148, 722 (1966).
- 40 S.K. Lyo and E.D. Jones, Phys. Rev. B 38, 4113 (1988).
- ⁴¹ A. Pinczuk, J. Shah, H.L. Störmer, R.C. Miller, A.C. Gossard, and W. Wiegmann, Surf. Sci. 142, 492 (1984).
- ⁴²H. Buhmann, Ph.D. thesis, Konstanz, Germany, 1992.

Phys. Rev. B 40, 9772 (1989).

 27 H. Kalt (unpublished).