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Influence of degenerate free carriers on radiative lifetimes in GaAs quantum wells

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The influence of the Coulomb interaction on the radiative recombination rate of minority photocreated carriers in a GaAs quantum well containing a degenerate free-electron or hole gas at 1.8 K is investigated using time-resolved photoluminescence. A factor of ≈ 1.5 enhancement in the recombination rate compared to that expected without the Coulomb interaction is observed experimentally at a carrier density of 8×10^{10} cm^{-2}. The recombination rate is also calculated theoretically as a function of carrier density and minority carrier localization in the presence of the Coulomb interaction. An enhanced rate is obtained due to correlation between the degenerate Fermi gas and the minority carrier at the Fermi edge, but only when the minority carrier is localized. The degree of localization required to reproduce the experimental data is determined.

I. INTRODUCTION

The photoluminescence (PL) spectra of undoped quantum-well (QW) structures at low temperature are normally dominated by the radiative recombination of two-dimensional (2D) Wannier excitons. However, if a high-density degenerate electron or hole gas is introduced into a QW by doping or electric-field injection, recombination takes place between the photocreated minority carrier and the degenerate free-carrier gas (DFG). Many-body Coulomb correlation between the DFG and the minority photocreated carrier causes enhancement of absorption at the Fermi edge giving rise to the so-called Fermi-edge singularity (FES) resonances in photoluminescence excitation (PLE) spectra.[1,2] If the minority carrier is localized, the FES can also appear in PL spectra. Localization spreads the minority carrier wave function in k space, allowing transitions near the Fermi edge.[3,4]

The complicated evolution of the absorption $A(\omega)$ and emission $E(\omega)$ spectrum as a function of frequency ω and degenerate electron density in such samples has been recently discussed theoretically.[5] It has been shown that while a complicated line shape results due to a combination of excitonic and shake-up effects, the total integrated intensity is relatively simple to understand.[5-7] The matrix elements which give the total intensity, and also the radiative lifetime, are given by the overlap between the initial many-electron wave function and the wave function perturbed by the presence of the valence hole. Hence, the radiative lifetime is a direct measurement of correlations in a degenerate free-carrier gas induced by a minority carrier.

There has been, however, little direct theoretical or experimental work which studies quantitatively the effect of this correlation on the radiative lifetime in a quantum well containing a degenerate free-carrier gas. In the absence of electron-hole correlation and if the temperature is sufficiently low that the electron or hole gas remains degenerate, the radiative lifetime of photocreated minority carriers at $k=0$ would be determined only by the momentum matrix element p_{vc} for interband transitions, and the electron-hole envelope-function overlap integral.[8] This high carrier density regime has been investigated by Matsusue and Sakaki[8] using a heavily n-doped (10^{12} cm^{-2}) 9.6-nm GaAs multiple QW. They obtained a somewhat shorter lifetime (440 ps at 15 K) than expected from a calculation based on the magnitude of the momentum matrix element expected from $\mathbf{k}\cdot\mathbf{p}$ theory for transitions at $k=0$ (560 ps), assuming no Coulomb correlation between the photocreated hole and the degenerate sea of electrons. This calculated value of the recombination time without Coulomb interaction (which we call here the "noninteracting lifetime" τ_0) is in agreement with other calculations.[9]

Recently, Skolnick *et al.*[10] estimated in a modulation-doped $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$ strained-layer quantum well an enhancement of a factor of 2 in a radiative recombination rate for a minority hole transition to an $n=2$ electron subband containing a low-density degenerate electron gas which exhibits FES effects in PLE compared to the transition to the $n=1$ electron subband which contained a high density of carriers and no FES effects. This suggests that Coulomb correlation effects at the Fermi edge can significantly influence the radiative lifetime of minority carriers, at least at low carrier densities. This estimation was made, however, rather indirectly from cw PL measurements using a factor of 11 correction for the different envelope-function overlaps of the two transitions. The effect of a low density of free carriers on the optical properties of resonant tunneling diodes has been investigated by Young *et al.*[11]

Radiative recombination rates can be determined much more directly in the time domain using time-resolved photoluminescence (TRPL). In this paper, we report TRPL investigations of the radiative lifetime of minority carriers in a GaAs quantum well containing a DFG. We present complementary theoretical calculations of the effect of the Coulomb interaction on the lifetime as a function of carrier density and localization, and compare experiment and theory. We study a quantum-well structure where external biasing of a Schottky barrier contact is used to inject a degenerate electron or hole gas into the quantum wells. The transformation from free Wannier exciton luminescence to luminescence in the presence of degenerate free carriers can therefore be monitored directly in the same sample. The strong effects of electron-hole (FES) correlations in this sample have been previously investigated in time-integrated PL and PLE measurements[12] and magneto-PLE measurements.[13]

II. EXPERIMENT

Two GaAs quantum wells of width 10 and 20 nm separated and bounded by three 31-nm $Al_{0.3}Ga_{0.7}As$ barriers were investigated. The quantum wells were separated from the GaAs n^+ (100) substrate by a 500-nm undoped GaAs layer. An electric field was applied perpendicular to the plane of the quantum wells by means of a semitransparent 3-mm-diameter sputtered indium-tin-oxide Schottky barrier contact at the sample surface. On application of a bias voltage, a degenerate electron gas (positive bias) or hole gas (negative bias) was injected into the quantum wells. This injection process is attributed to impurity-assisted barrier tunneling. A more detailed discussion of the transport mechanism is given elsewhere.[12] The presence of the degenerate carriers was indicated by a bias voltage dependent increase in the Stokes shift (the difference between PL and PLE peak energy positions) as shown in Figs. 1 (10-nm QW) and 2 (20-nm QW). Figure 3 shows examples of the PL and PLE spectra (taken for the 10-nm QW) from which the data in Figs. 1 and 2 were obtained. The arrows indicate the bias corresponding to zero electric field: this was determined from the minimum quantum-confined Stark shift in the PL transition energy. The Stokes shift ΔE_S increases due to blocking of absorption close to the band edge by the degenerate carriers, causing a Moss-Burstein shift in the absorption edge determined by the PLE measurement. The position of the PL peak is, however, predominantly uninfluenced by state filling and recombination is still due to electrons and holes in many-body states near the band edge. From the relationship $\Delta E_S=\pi\hbar^2N_c(m_e^{-1}+m_h^{-1})$ [and using $m_e=0.067m_0$ and $m_h=0.19m_0$ (Ref. 14)], we calculate the maximum injected carrier densities N_c of 8×10^{10} cm^{-2} for the 10-nm QW and $N_c=1.8\times10^{10}$ cm^{-2} for the 20-nm QW. The Stokes shift at zero field originating from the usual mechanism of spectral diffusion to the low-energy side of the absorption line prior to recombination was assumed to be constant with field and was subtracted out in this calculation. At very high fields the carriers are swept out of the quantum wells leading to a reduction in the Stokes shift at high biases.

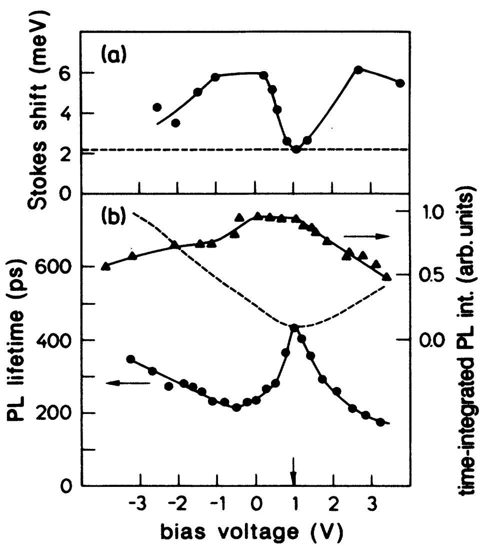

FIG. 1. (a) Stokes shift and (b) PL lifetime (dots) and PL efficiency (triangles) vs bias voltages for the 10-nm QW. Arrow: position of zero electric field. Dotted line: expected lifetime enhancement due to field-induced charge separation alone, as described in the text.

TRPL measurements were performed with the sample

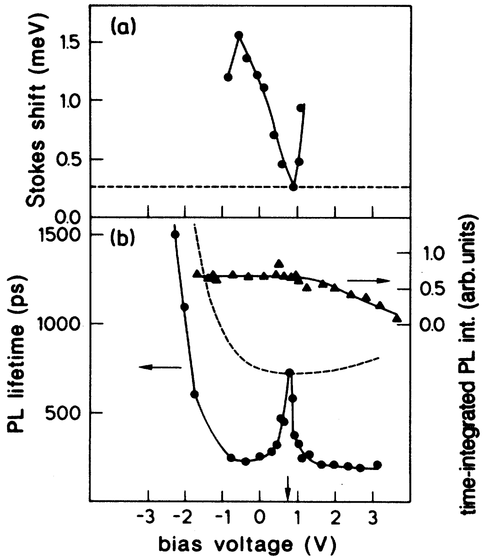

FIG. 2. (a) Stokes shift and (b) PL lifetime (dots) and PL efficiency (triangles) vs bias voltage for the 20-nm QW. Arrow: position of zero electric field. Dotted line: expected lifetime enhancement due to field-induced charge separation alone, as described in the text.

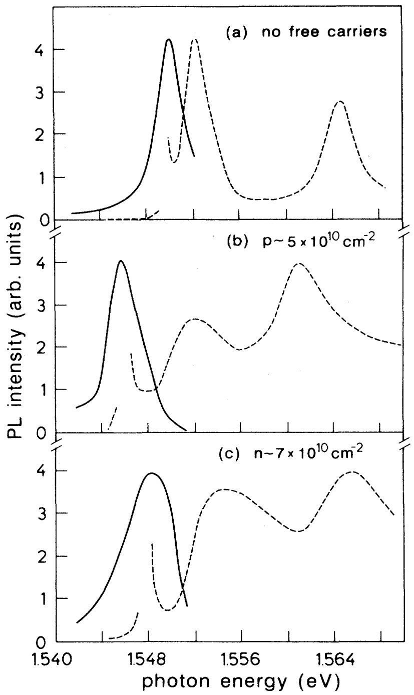

FIG. 3. PL spectra (full lines) and PLE spectra (dashed lines) for the 10-nm QW for different degenerate free-carrier densities at 1.8 K, (a) no free carriers, (b) hole density 5×10^{10} cm^{-2}, (c) electron density 7×10^{10} cm^{-2}.

held at a temperature of 1.8 K in a liquid-helium cryostat. The sample was excited with 2-ps pulses from a synchronously-pumped mode-locked Styryl 8 dye laser operating at 735 nm. The excited electron-hole pair carrier density was 1×10^{9} cm^{-2} per pulse. The PL was spectrally dispersed in a $\frac{1}{4}$-m monochromator and temporally dispersed in a synchroscan Delli-Delti DS/S streak camera with a low-frequency scanning photomultiplier readout system.[15] The temporal resolution was varied between 25 and 50 ps dependent on the required time resolution. The luminescence from the sample exhibited a finite rise time on a time scale of 100–200 ps due to hot-carrier cooling[16,17] followed by an exponential decay which could be followed over between one and three orders of magnitude depending on the magnitude of the PL lifetime compared to the streak camera scan period. Observation of an exponential decay indicates that excited carriers are close to thermal equilibrium with the lattice.[8] No significant spectral variation of the PL decay time was found across the luminescence line.

Figures 1 and 2 also show the PL decay time $\tau_{\rm PL}$ in the two QW's as a function of bias. We find a pronounced decrease in $\tau_{\rm PL}$ for both wells upon injection of the DFG. To determine the origin of this effect we also carefully monitored the time and spectrally integrated PL efficiency η of the PL transition in each quantum well. A reduction in PL lifetime could be due to an increase in nonradiative recombination due to extraction of carriers from the well by tunneling or leakage. However, as shown in Figs. 1 and 2, for negative fields the PL efficiency remains constant over the field regime where the PL lifetime falls. Most dramatically, for the 20-nm well at negative fields we observe a factor of 3 reduction in $\tau_{\rm PL}$ without any significant change in the PL efficiency of the QW. The PL efficiency data therefore indicate that the fall in PL lifetime for negative field is not due to nonradiative recombination. In addition, this indicates that the maximum PL efficiency corresponds to 100%, because in the presence of nonradiative recombination a change in PL lifetime must be accompanied by a change in observed PL efficiency if either the radiative or nonradiative lifetime changes. Assuming a peak PL efficiency of 100%, the positive field PL lifetimes in both wells can be easily corrected for nonradiative recombination to obtain the true radiative lifetime ($\tau_{\rm rad}=\tau_{\rm PL}/\eta$). The reduction in PL efficiency was found to be insufficient to explain the magnitude of the decrease in $\tau_{\rm PL}$ for either well. The PL efficiency measurements therefore show that, in both QW's for either bias, the reduction in PL lifetime with field is not due to nonradiative effects.

It is therefore clear that the injection of the degenerate carriers leads to a reduction in the radiation lifetime in these samples. For both wells a minimum lifetime of around 200 ps is observed. At higher negative bias we observe an increase in $\tau_{\rm PL}$ due to reduced overlap of the electron and hole envelope functions[18,19] induced by the field. No recovery of $\tau_{\rm PL}$ is observed for positive fields due to the larger decrease in PL efficiency. Figures 1 and 2 also show (dotted line) the calculated increase in $\tau_{\rm PL}$ expected with field due to this spatial separation of the electron and hole alone. The change in the electron-hole overlap integral as a function of field was obtained from an envelope-function calculation as described in Ref. 19. The electric field across the well was calculated from the measured quantum-confined Stark shift in the PL line position at each bias. This field enhancement calculation was used to correct the PL lifetimes to zero field, and the PL efficiency was used to correct for nonradiative effects as described earlier.[20] After this procedure, we obtained from Figs. 1 and 2 the variation of minority carrier radiative lifetime versus majority carrier density shown in Fig. 4. Although the 20-nm quantum well exhibits a somewhat larger fall in PL lifetime for the degenerate hole

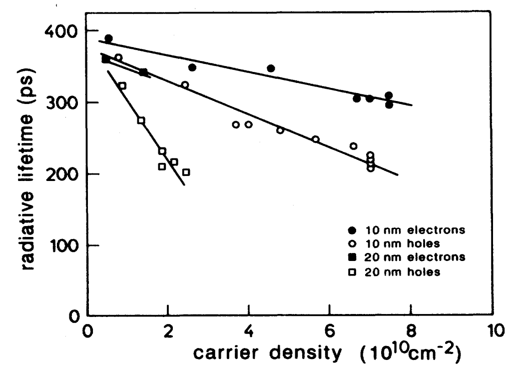

FIG. 4. Radiative lifetime vs carrier density obtained from Figs. 1 and 2, after correcting for effects of nonradiative recombination and reduced electron-hole overlap. For each case, the carrier types in the legend refer to the majority injected carrier type.

case, it is clear that a radiative lifetime of 200–400 ps is obtained in this carrier density regime. Such values are factors of 1.5–2.8 less than calculated in Refs. 8 and 9 in the absence of the Coulomb interaction.

III. THEORY

In order to interpret these observations, we now present a theoretical calculation of the reduction in minority carrier lifetime due to Coulomb correlation with a Fermi sea. It has been generally accepted that localization of the minority carrier is required to observe enhanced recombination near the Fermi edge in cw PL spectra. We therefore consider the case of a photocreated minority hole of arbitrary localization radius in the presence of a degenerate electron gas.

First, however, we discuss the additional influence of the localization on the minority free-carrier lifetime due to momentum conservation even in the absence of Coulomb effects. A delocalized hole in its lowest-energy state occupies a well-defined k state at $k=0$. By momentum conservation, recombination can only take place with $k=0$ electrons because the emitted photon carries away negligible momentum. The radiative lifetime is then given by the noninteracting (density-independent) degenerate lifetime τ_0 as illustrated in Fig. 5(a). Localization spreads the hole in k space, allowing recombination with $k>0$ electron states. In the case where the localization is not sufficiently strong to spread the hole wave function further than k_F, the oscillator strength contributing to τ_0 in the delocalized hole transition of Fig. 4(a) becomes spread among all the transitions now allowed with the localized hole [Fig. 5(b)]. Assuming p_{cv} is independent of k (and no Coulomb interaction), summing all the transition probabilities gives again a lifetime of τ_0.

However, if the localization spreads the hole far beyond k_F, k components of the hole wave function above k_F see unoccupied electron states [Fig. 5(c)]. A lifetime of roughly $[1+(k_{\text{loc}}/k_F)^2]\tau_0$ is then expected. Localization, even in the absence of the Coulomb interaction, can therefore modify the radiative lifetime of the hole, leading to a radiative lifetime comparable to τ_0 for $(k_{\text{loc}}/k_F)<1$, but greater than τ_0 for $(k_{\text{loc}}/k_F)>1$. These momentum conservation effects must be considered when determining the absolute radiative lifetime of the hole.

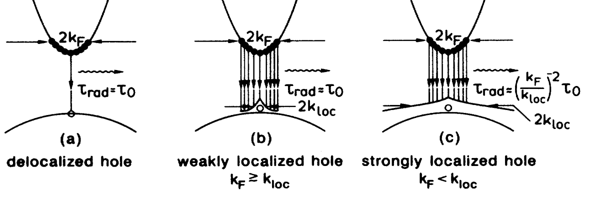

FIG. 5. Localization of a hole in the presence of a degenerate Fermi sea of electrons increases the radiative lifetime compared to a delocalized hole τ_0 by momentum conservation if $k_{\text{loc}} \gg k_F$. See the text for further details.

We now consider the influence of the Coulomb interaction on the radiative recombination rate. Since the excited minority carrier density in our experiments is much less than the peak degenerate carrier density, we consider the ideal case of the correlation of the many carriers in the Fermi sea with a single minority carrier. We initially assume the minority carrier is a photocreated hole which has energetically relaxed and become localized, e.g., in a well width fluctuation island, of arbitrary width. The localization is described by applying a Gaussian envelope function of width a_{loc} ($\approx 1/k_{\text{loc}}$), to the hole wave function in the plane of the quantum well.

The radiative lifetime τ_{rad} is obtained from the sum of the matrix elements over all occupied electron states $|\lambda\rangle$ (i.e., all the states with energies e_λ, less than the Fermi energy E_F),

$$1/\tau_{\text{rad}} = \sum_{e_\lambda < E_F} m_\lambda^2 \ . \tag{1}$$

The matrix elements m_λ can be expressed as a sum of terms given by the product of matrix elements $m_k = p_{cv}\langle k|h\rangle$ for transitions in the absence of the Coulomb interaction (where $|h\rangle$ is the localized hole state), and the overlap between free electron states $\langle k|$ and electron states $|\lambda\rangle$ in the presence of the hole potential, i.e.,

$$m_\lambda = \sum_k m_k \langle k|\lambda\rangle \ . \tag{2}$$

For a constant matrix element m_k, these elements are just the probability of finding an electron at the position of the hole. The overlap integrals are solutions of the Wannier equation,

$$e_k\langle k|\lambda\rangle + \sum_{k'} V_{k,k'}\langle k'|\lambda\rangle = e_\lambda\langle k|\lambda\rangle \ . \tag{3}$$

The interaction $V_{k,k'}$ is the screened attractive potential of the hole. The solutions of Eq. (3) and the matrix elements m_λ are discussed in detail in Refs. 5 and 6.

Using this formalism we calculated the ratio of carrier lifetime when the minority carriers interact Coulombically with the Fermi sea (τ_{int}), to the carrier lifetime obtained by the same method but for zero Coulomb interaction (τ_{nonint}). Figure 6 shows this ratio as a function of carrier density for different localization radii relative to the 3D GaAs exciton Bohr radius a_B. By using $\tau_{\text{int}}/\tau_{\text{nonint}}$, we eliminate the effects of momentum conservation on the absolute radiative lifetime shown in Fig. 5 (we assume these effects to be independent of the presence of the Coulomb interaction). This ratio, therefore, conveniently allows the effect of the Coulomb interaction on the radiative lifetime to be examined alone. At $N_c = 8\times10^{10}$ cm^{-2} (the peak carrier density achieved in our experiment), $\tau_{\text{int}}/\tau_{\text{nonint}}$ varies with $k_F a_{\text{loc}}$ as shown in Fig. 7. The results in Figs. 6 and 7 show that a Coulomb enhancement of the recombination rate occurs only when the localization spreads the hole wave function as far as k_F. The enhancement therefore comes mainly from states in the vicinity of the Fermi energy and not significantly from Coulomb correlation between the hole and electrons near $k=0$. For $k_F \ll 1/a_{\text{loc}}$, we find that

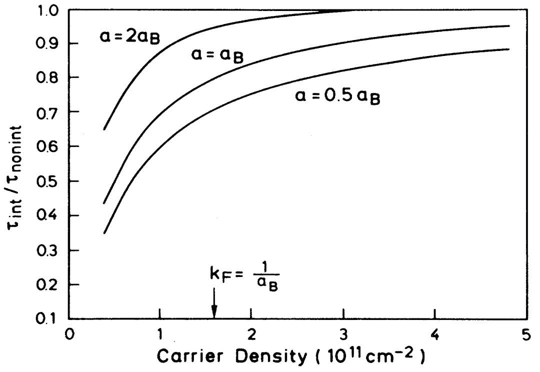

FIG. 6. $\tau_{\rm int}/\tau_{\rm nonint}$ vs degenerate electron density N_c for three different hole localization radii $a_{\rm loc}$.

$\tau_{\rm int}=0.5\tau_{\rm nonint}$. For $k_F \approx 1/a_{\rm loc}$, a radiative lifetime of approximately $0.7\tau_{\rm nonint}$ is obtained. However, for $k_F \gg 1/a_{\rm loc}$, negligible enhancement in radiative recombination rate occurs.

IV. DISCUSSION

In order to compare with the absolute lifetimes obtained in measurements, we combine these results with the momentum-conservation arguments given in Fig. 5. For $N_c=8\times10^{10}$ cm^{-2}, we find from Fig. 6 that if the minority carriers become localized to an area of radius a_B, then a lifetime $\approx 06\tau_{\rm nonint}$ is expected, close to the experimental value for a DFG of electrons *if* $\tau_{\rm nonint}=\tau_0\approx560$ ps. At $N_c=8\times10^{10}$ cm^{-2} and $a_{\rm loc}=a_B$, $k_F=0.7/a_{\rm loc}\approx0.7k_{\rm loc}$. A large increase in lifetime is therefore not expected due to the momentum-conservation effects, so the condition $\tau_{\rm nonint}\cong\tau_0$ is indeed satisfied. The trend to larger lifetimes at smaller N_c is explicable by the influence of the momentum-conservation effects, since k_F is then somewhat less than $k_{\rm loc}$ (see Fig. 5). Considering the simplicity of the model used, we can conclude that the experimental data are roughly consistent with a localization of the minority carrier within an area πa_B^2. Note that the theoretical results show that enhanced radiative recombination would not normally be expected at carrier densities greater than $N_c=5\times10^{11}$ cm^{-2} because very strong localization of the minority carriers would be required. This is consistent with the lifetime close to τ_0 observed in Ref. 8 at $N_c=1\times10^{12}$ cm^{-2}, and the recombination enhancement of a factor of 2 between small and large N_c subbands observed in Ref. 10. Our findings, therefore, seem consistent with other experimental studies. The minority carrier density dependence of radiative and nonradiative recombination rates has also been studied in a GaAs/Al$_x$Ga$_{1-x}$As modulation-doped quantum well by Liu *et al.*,[21] where only a small decrease in radiative recombination rate was observed between 0 and 1×10^{11} electrons/cm^{-2}. We believe this result may be due to the much larger minority photoexcited carrier density ($\leq5\times10^{10}$ cm^{-2}) used, leading to a saturation of any available localization sites, and perhaps also heating of the DFG.

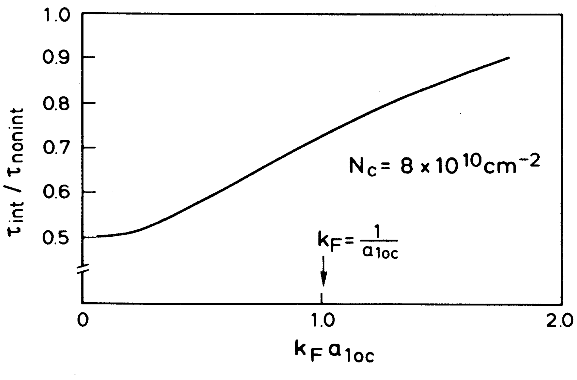

FIG. 7. $\tau_{\rm int}/\tau_{\rm nonint}$ vs $k_F a_{\rm loc}$ at $N_c=8\times10^{10}$ cm^{-2}.

We now discuss an intrinsic mechanism to supply the broadening of the minority carrier in k space without the need to postulate a certain form of interface roughness. In the above calculation, we chose for convenience to consider a fixed minority carrier localized to certain area (inclusion of minority carrier motion leads to an extremely complex many-body treatment). However it is clear that even for a quantum well containing zero interface roughness, a pure $k=0$ minority carrier Bloch wave extending throughout the crystal would still not be obtained. Rapid scattering between, for example, a minority hole and those degenerate electrons within k_BT of E_F (and therefore available for scattering) would cause rapid momentum relaxation of the hole. These active electrons correspond to a scattering density of $\approx4\times10^9$ electrons/cm^2 at any E_F (i.e., at any two-dimensional electron gas density in the degenerate limit) at 1.8 K.

This scattering would severely limit the coherence length of any hole k state, and mix higher k states into the hole wave function in the same way as a localization potential. Free-carrier dephasing is believed to be a very efficient process,[22] occurring on a subpicosecond time scale. A dephasing time of 1 ps corresponds to a homogeneous linewidth of 1.37 meV,[22] and thus to an uncertainty in the hole k vector of 7.9×10^7 m^{-1}. Since at $N_c=8\times10^{10}$ cm^{-2}, $k_F=7\times10^7$ m^{-1}, it seems likely that carrier-carrier scattering could provide sufficient broadening of the hole k states to allow interaction with the Fermi edge even in the absence of any localization by crystal imperfections, at least for $N_c\leq8\times10^{10}$ cm^{-2}. In this case, we would expect our observations to be rather general and not dependent on the interface morphology or defect concentration of a specific sample. However, we would still expect the fixed localized hole calculation to provide a reasonable value of $\tau_{\rm int}/\tau_{\rm nonint}$ at a given hole coherence length since the broadening of the k vector of the hole still dominates the hole motion. It is worth noting that a similar mechanism affects the lifetime of atomic Wannier excitons in quantum wells.[23–26] Limitation of the coherence length of the exciton center-of-mass motion by scattering causes a mixing of higher exciton **K** states into the **K**=0 exciton state. Because of momentum conservation, higher exciton **K** states are not optically active.[23] The Wannier exciton lifetime even at very low temperature is therefore greatly *increased* above the expected **K**=0 value ($\approx$25 ps) by finite coherence length.[24,26] However, in the case of a degenerate Fermi gas, the $k > 0$ minority carrier states remain optically active due to the occupancy of $k > 0$ majority carrier states. The minority carrier lifetime therefore does not increase but, as demonstrated above, can decrease due to enhanced recombination probability near the Fermi edge. The existence of a finite Wannier exciton coherence length and optically active $k > 0$ degenerate carrier states explains why we obtain the perhaps rather counterintuitive experimental result (Figs. 1 and 2) that the radiative recombination rate of the highly correlated electron-hole pair in a Wannier exciton can be lower than that of a minority carrier in the presence of a degenerate free-carrier gas.

V. CONCLUSIONS

We have studied the influence of degenerate free carriers on the radiative lifetime of a minority photocreated carrier in a quantum well at 1.8 K both experimentally and theoretically. For $N_c \approx 8 \times 10^{10}$ cm^{-2}, experimentally determined lifetimes are shown to be $\approx 60\%$ of those expected without the Coulomb interaction, and shorter than the Wannier exciton lifetime due to the reduced coherence length of the Wannier exciton state. $\tau_{\rm rad}$ is calculated theoretically as a function of carrier density and localization area. Significant enhancement of the radiative lifetime occurs only when localization of the minority carrier introduces k components near k_F. The experimental enhancement was found to be consistent with localization in an area of $\approx \pi a_B^2$. Larger N_c would be expected to lead to much weaker radiative enhancement due to the requirement of a much stronger localization. Finally, we propose that the spread of the minority carrier in k space could be accomplished intrinsically by a large reduction in the minority carrier coherence length due to carrier-carrier scattering.

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