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Long-lived spatially indirect excitons in coupled $GaAs/Al_xGa_{1-x}As$ quantum wells

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We present nanosecond time-resolved measurements of the low-temperature photoluminescence in coupled $GaAs/Al_xGa_{1-x}As$ quantum wells. Working with 10.0-nm and 15.0-nm GaAs quantum wells coupled through a 3.0-nm $Al_{0.3}Ga_{0.7}As$ barrier layer, we observe spatially indirect excitons with lifetimes in excess of 200 nsec—an enhancement of more than 2 orders of magnitude over the lifetime of the spatially direct exciton. As a result, extremely cold GaAs exciton gases become possible, a prerequisite for testing recent predictions of quantum-statistical behavior in this system.

INTRODUCTION

There is a growing body of literature on the optical properties of coupled quantum wells—two low-band-gap well layers separated by a thin, high-band-gap barrier layer.[1-12] Coupled wells have been studied for their interesting spectroscopy,[1-6] as paradigms for more complicated tunneling structures,[7-9] and for applications to optoelectronic devices.[10-12]

A number of workers have recently considered spatially indirect excitations in coupled wells.[4,13-15] These excitations—consisting of an electron and hole confined to different quantum-well layers—have properties which differ dramatically from those of spatially direct, single-quantum-well excitons. A particular example is the greatly enhanced recombination lifetime which should accompany the separation in space of electron and hole. Lifetime enhancements as great as 10^5 have been predicted in idealized systems.[14]

There are a number of motivations to study such excitations. First, the possibility to tune the recombination lifetime of excitons over orders of magnitude in a material consisting entirely of direct-gap semiconductors is an interesting test of modern band-gap engineering and crystal-growth techniques. Second, we wish to evaluate the applicability of the coupled $GaAs/Al_xGa_{1-x}As$ quantum-well system to a recent work on the possibility of Bose condensation[14] and to a series of older works on excitonic superconductivity.[16,17] Specifically, if spatially indirect GaAs excitons are to provide a testing ground for these theories, we must show that these excitations can persist for a time long enough to come to thermal equilibrium with the lattice—about 10 nsec for GaAs at 4 K.[18]

Our work differs in a number of ways from the recent work of Charbonneau *et al.*[13] Most importantly, we report lifetimes more than 2 orders of magnitude longer than the 1.9-nsec result presented in Ref. 13. This result is crucial if we are to study the high-density behavior of a gas of *cold* excitons. Also, we work with an asymmetric coupled-well system. This design permits us to contrast the response of the spatially direct, single-well exciton with that of the spatially indirect, double-well exciton.

We present the results of temporally and spectrally resolved measurements of photoluminescence in coupled $GaAs/Al_xGa_{1-x}As$ quantum wells. The main results of these measurements are as follows. (i) Spatially indirect excitons with very great lifetime enhancements can be produced. The observed decay is nonexponential and cannot be characterized by a simple lifetime. However, the spatially indirect excitons are exceedingly long lived with a significant fraction of the initial population persisting in excess of 1 μsec. This decay is more than 2 orders of magnitude slower than the radiative lifetime of GaAs single-quantum-well excitons. (ii) Fluctuations in the barrier width give rise to a distribution of spatially indirect exciton decay times. (iii) It is possible to produce a gas of fully thermalized, spatially indirect excitons, perhaps even at temperatures below 1 K. This result contrasts with the situation for single-quantum-well excitons where recombination times are shorter than the tens of nanoseconds required for complete thermalization.

EXPERIMENT

The sample was grown by molecular-beam epitaxy on a GaAs:Si[001] substrate. After buffer layers, we grew a 1000-nm $Al_{0.3}Ga_{0.7}As$:Si contact layer, a 400-nm $Al_{0.3}Ga_{0.7}As$ spacer layer, a short-period-superlattice smoothing sequence, a 10.0-nm GaAs well layer, a 3.0-nm $Al_xGa_{1-x}As$ barrier layer, a 15.0-nm GaAs well layer, a 500-nm $Al_{0.3}Ga_{0.7}As$ spacer, a 1000-nm $Al_{0.3}Ga_{0.7}As$:Be contact layer, and a 10-nm GaAs:Be cap. Doping concentrations were nominally 1×10^{18} cm^{-3}. The sample was patterned into mesas of area 2×10^{-3} cm^{-2} using conventional photolithographic techniques, and ring-shaped Ohmic contacts were fabricated. The sample was mounted on a sapphire substrate and affixed to the cold finger of a flowing helium cryostat. The ultimate temperature, measured at the cold finger, was 5.6 K. The built-in potential of the *p-i-n* diode at this temperature was determined to be $V_{bi}=1.55$ V by observations of the photocurrent yield.

The sample was excited using a synchronously pumped rhodamine-6G laser tuned to 590 nm and cavity dumped at 40 kHz. The excitation pulse duration was 5 psec. The luminescence was dispersed through a 1-m double spectrometer and detected using a cooled photomultiplier operated in photon-counting mode. Temporal resolution

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of 0.4 nsec ($1/e$ response) was achieved using a constant fraction discriminator and time-to-amplitude converter. Time decays were recorded using a multichannel analyzer. Time-resolved spectra were recorded using a single-channel analyzer and scanning the spectrometer.

RESULTS—FLAT BAND

To study the spatially direct system, we forward-biased the *p-i-n* diode to partially cancel the built-in potential. The results are shown in Fig. 1. Under these near-flat band conditions ($V_{bias} = +0.9$ V), the time-integrated photoluminescence spectrum consists of a strong peak at 809.25 nm and a much weaker emission at 805.7 nm. Comparison with a one-band, effective-mass calculation, and studies of the electric-field dependence of these lines, permit us to identify these lines as heavy-hole (809.25 nm) and light-hole (805.7 nm) excitation emissions arising from the 15.0-nm quantum well. The light-hole emission plays no role in the following and will not be discussed further. The decay of the heavy-hole excitonic emission following pulsed excitation is exponential over two *e*-folding times with ($1/e$) decay time 1.5 ± 0.2 nsec. When deconvolved with the system response of 0.4 nsec, this implies a lifetime of 1.4 ± 0.2 nsec.

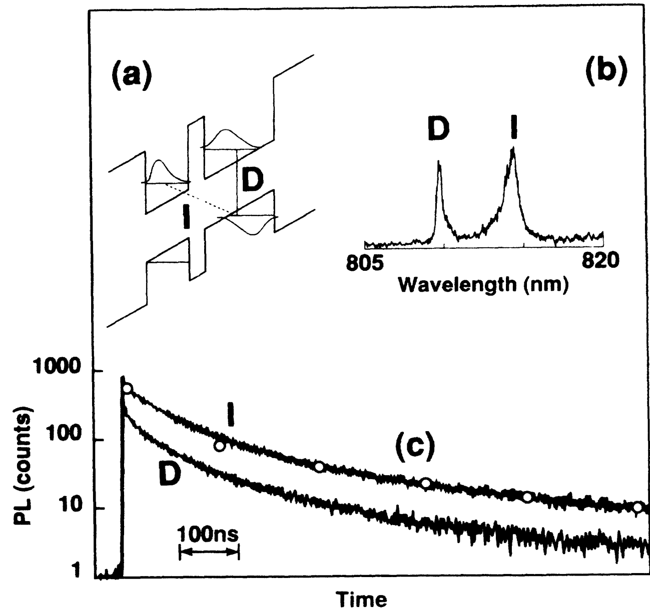

FIG. 2. (a) Band diagram showing direct (D) and indirect (I) exciton transitions. (b) Time-integrated PL spectrum showing the direct exciton emission at 810.55 nm and the indirect emission at 815.85 nm for $V_{bias} = -0.2$ V. (c) Time decays of the direct and indirect emissions following pulsed excitation. The open circles are theory.

RESULTS—REVERSE BIAS

To study the spatially indirect system, we reverse-biased the *p-i-n* diode. The results are shown in Fig. 2. The time-integrated photoluminescence spectrum for $V_{bias} = -0.2$ V shows clearly the presence of sharp and well-resolved peaks at 810.55 and 815.85 nm. Comparison with a one-band effective-mass calculation, and studies of the electric-field dependence permit us to identify the 810.55-nm line as the spatially direct heavy-hole exciton emission originating in the 15.0-nm quantum well (e_2-hh_1 emission). The 815.85-nm line is due to the recombination of the spatially indirect exciton consisting of a heavy-hole confined to the 15.0-nm well layer, and an electron confined to the 10.0-nm well layer (e_1-hh_1 emission).

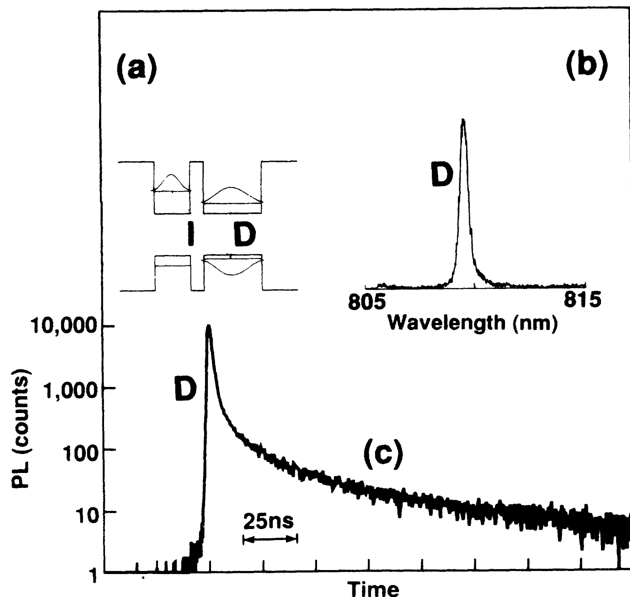

FIG. 1. (a) Band diagram showing direct (D) and indirect (I) exciton transitions. (b) Time-integrated PL spectrum showing the direct exciton emission at 809.25 nm for $V_{bias} = +0.9$ V. (c) Time decay of 809.25-nm emission following pulsed excitation.

Figure 2(c) shows the time decay of both the 810.55- and 815.85-nm lines. The 810.55-nm line (spatially direct exciton) shows an initial fast decay ($1/e$ time: 1.4 ± 0.2 nsec after deconvolving instrumental response) followed by a slow, nonexponential tail. The 815.85-nm line exhibits a much less prominent initial spike, and then a long-lived tail. Careful examination of the data shows that after the initial transient, the two lines follow the same decay.

We took several steps to confirm that this long-lived, nonexponential feature is due to intrinsic emission by carriers pumped into the coupled wells instantaneously with the excitation pulse. In order to rule out the possibility that the emission is pumped over a microsecond time scale by the capture of slow carriers from the $Al_xGa_{1-x}As$ layers, we time-resolved the strong photoluminescent emission originating in the $Al_xGa_{1-x}As$ itself. The time decay essentially reproduces our detection-system response and we conclude that carriers are depleted from the $Al_xGa_{1-x}As$ layers on a subnanosecond time scale. We found this result to hold over the range of bias voltages used in this study. Simple calculations and WKB esti-

mates of the tunneling rates,[19] as well as recent experiments,[7-9] show that electron- and hole-tunneling times through the 3.0-nm barrier are subnanosecond leading to fast relaxation of hot carriers. Calculations of carrier cooling by acoustic phonon emission show that the electron gas equilibrates with the lattice in a time of order 10 nsec.[18] We therefore conclude that the long-lived emission shown in Fig. 1 results strictly from the dynamics of carriers captured into the wells essentially instantaneously with the laser-excitation pulse.

DISCUSSION

We seek to understand these results in terms of a simple picture of the dynamics of electrons and holes in a coupled quantum-well structure. Accordingly, we assume that electrons and holes are pumped into the wells instantaneously with the laser pulse. We ignore any imbalance in carrier concentration that may arise because of bias conditions or differing capture cross sections. We also ignore any differences in the capture into the 15.0- and 10.0-nm well layers.

For flat-band conditions electrons and holes form spatially direct excitons in the 15.0-nm quantum well in approximately 0.4 nsec and begin to recombine.[20] The lifetime for the radiative recombination can be estimated from the results of Ref. 21 as $\tau = 1.9$ nsec. The decay of the 809.25-nm feature is characterized by a lifetime of 1.4 nsec, slightly faster than the pure radiative rate we estimate. We conclude that the fast decay reflects spatially direct recombination in the 15.0-nm quantum well, and that the recombination is substantially radiative in nature.

For reverse-bias conditions, a simple rate analysis gives the following results: (i) The fast spike evident during the first few nanoseconds in Fig. 2(c) is due to the spatially direct recombination of e_1-hh_2, e_2-hh_1 excitons, and to the relaxation of hot electrons from $|e_2\rangle$ to $|e_1\rangle$ and of hot holes from $|hh_2\rangle$ to $|hh_1\rangle$. This interpretation was supported by an observation of a luminescence signal at the e_1-hh_2 energy with $\approx$ 1-nsec decay time. (ii) The spatially direct (e_2-hh_1) recombination mirrors the indirect decay because of the thermal excitation of population from $|e_1\rangle$ to $|e_2\rangle$. This interpretation is confirmed by measurements of the temperature dependence of the time-integrated photoluminescence spectrum. The results show that the direct recombination is thermally activated. (iii) The nonexponential character of the observed decay is not predicted in the present rate model.

In order to explain the nonexponential nature of the luminescence decay, we must introduce a distribution of decay times. Such a distribution arises as a result of interfacial roughness and the interplay between the localized and delocalized exciton populations. We first consider the localized component. Localized excitons experience a distribution of decay times as a result of barrier-thickness fluctuations. We model the effect by assuming that the barrier thickness is distributed according to

$$g(\xi) = \frac{\alpha}{\sqrt{\pi}\xi} \exp\{-[\alpha(L_B - L_{B0})/\xi]^2\} , \quad (1)$$

where L_{B0} is the nominal $Al_xGa_{1-x}As$ barrier thickness, $\alpha^2 = 4\ln 2 = 2.77$, and ξ is the full width at half maximum of the distribution. In choosing ξ, we assume $\pm$ half-monolayer fluctuations in the normal interface ($Al_xGa_{1-x}As$ on GaAs) and $\pm$ monolayer fluctuations in the inverted interface (GaAs on $Al_xGa_{1-x}As$). We further assume the roughness at the two interfaces in uncorrelated, and compute $\xi = [(1\ \mathrm{ML})^2 + (2\ \mathrm{ML})^2]^{1/2} = 0.63$ nm, where ML = 0.283-nm represents the monolayer thickness. For each value of L_B, we then compute the recombination lifetime as $\tau = \tau_D |\langle e_1 | hh_1 \rangle|^2$, where the wave functions are computed for each barrier thickness in a simple one-band effective-mass scheme, and τ_D is the radiative recombination rate under spatially direct conditions. τ_D is taken equal to the spatially direct recombination rate reported above, i.e., 1.4 nsec. This simple model is consistent with the observed time-integrated photoluminescence linewidths.

Mobile excitons do not experience a static distribution of decay times. Instead, they sample the entire distribution as they migrate through the crystal. This process gives added weight to the fast recombination. In fact, since a locally thin barrier layer corresponds to a locally wide quantum well, excitations are driven to points of relatively rapid recombination. We therefore expect that mobile excitons will decay in a time comparable to the fastest recombination calculated using the static model presented above, i.e., about 50 nsec.

The relative weights of the localized and mobile components can be estimated from a simplified rate analysis based on Fig. 3. In Fig. 3, $|1\rangle$ ($|2\rangle$) represents a localized (mobile) exciton, i.e., one below (above) the mobility edge. Level $|0\rangle$ denotes the vacuum state of excitons. Levels $|1\rangle$ and $|2\rangle$ each decay by emitting photons at rates Γ_{loc} and Γ_{mob}, respectively. In addition, $|1\rangle$ and $|2\rangle$ are coupled by a rate w for spontaneous deexcitation. The corresponding rate for spontaneous excitation is obtained from detailed balance as $w\exp(-\Delta/kT)$, where Δ denotes the $|1\rangle$-$|2\rangle$ splitting. Here, Δ is set equal to the localization energy, approximately 2 meV in this system. The rate w is determined by the electron cooling rate, and this was calculated in Ref. 15. At 10 K, we have $w = (5\ \mathrm{nsec})^{-1}$, and $w\exp(-\Delta/kT) = (55\ \mathrm{nsec})^{-1}$. Integration of the corresponding rate equations shows that the weights

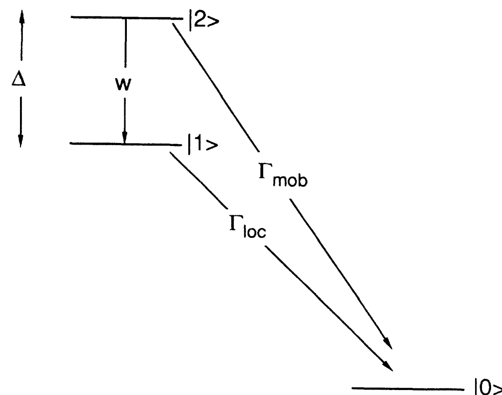

FIG. 3. Rate model used in computing the relative weights of the localized and mobile populations.

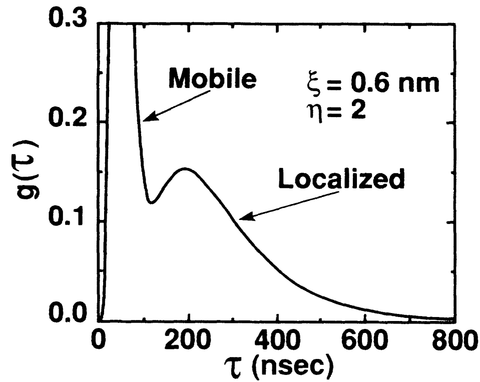

FIG. 4. Distribution of decay times calculated according to the model described in the text. The distribution is normalized to unity at the peak. $\xi = 0.6$ nm sets the barrier roughness through Eq. (1). $\eta = 2$ is the ratio of the mobile to localized components.

of the localized and mobile components are in the approximate ratio $\eta = 1{:}2$.

The total effective lifetime distribution, obtained as the weighted sum of the localized and mobile components, is that displayed in Fig. 4. The corresponding time decay is shown by the open circles in Fig. 2(c) and is seen to be in agreement with the observed response over the entire microsecond interval.

A number of alternative explanations for the nonexponential character of the photoluminescence (PL) decay suggest themselves, but may be ruled out. The kinetics of a slowly-cooling-electron distribution would yield a nonexponential dependence; however, calculations[18] of the electron cooling rates indicate that the decay must become exponential after approximately 10 nsec. Similarly, the process of exciton formation is nonexponential when comparable numbers of electrons and holes are present. However, this, too, must become exponential when a minority carrier is established, and in any event has been shown to be rapid in a similar system.[20]

CONCLUSIONS

We have contrasted the recombination dynamics of single-quantum-well excitons with those of spatially indirect excitons. The most striking difference reported here lies in the recombination lifetimes. While spatially direct excitons were observed to recombine on a 1-nsec time scale, a substantial fraction of indirect excitons persisted in excess of 1 μsec. This time scale is many times longer than the ≈ 10 nsec necessary for complete cooling and should permit studies of interesting quantum-statistical behavior in a gas of GaAs excitons. This exceedingly long lifetime is achieved in a crystal composed entirely of direct-gap semiconductors.

We have also analyzed the form of the photoluminescence decay. The observed nonexponential behavior cannot be explained by conventional mechanisms, but is in agreement with a model incorporating the effects of exciton localization and barrier-thickness fluctuations.

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