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Enhanced radiative decay in disordered GaAs/Al_{0.3}Ga_{0.7}As superlattices

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Characteristics of the photoluminescence (PL) from disordered and ordered GaAs/Al_{0.3}Ga_{0.7}As superlattices are compared. The disordered superlattices (DSL) are nonperiodic and the ordered superlattices are periodic. The PL emission is much stronger in the DSLs by a factor that depends on the PL excitation wavelength and excitation power. These observations are interpreted in terms of an enhanced radiative decay rate in the DSLs brought about by the breakdown of translational symmetry in these systems. © 1995 American Institute of Physics.

I. INTRODUCTION

The advent of semiconductor quantum-well structures has stimulated a heightened interest in the study of free excitons because free-exciton transitions dominate in these systems.^{1,2} The confinement of the kind produced in quantum wells leads to a large dipole moment for the excited electron and concomitant enhancement in the binding energy, oscillator strength, and radiative decay rate of the exciton. Large dipole moments lead to optical nonlinearity and suggest potential applications.³ Exciton radiative decay can be probed by photoluminescence (PL) experiments. In this work we have observed the enhancement of the radiative decay in systems where the exciton is localized by translational disorder, which is deliberately introduced in the form of nonperiodicity in one direction. We present results and analysis of PL measurements on periodic and nonperiodic GaAs/Al_{0.3}Ga_{0.7}As superlattices where the periodicity/nonperiodicity is in the growth direction of the superlattice. A periodic or ordered superlattice (OSL) is fabricated by repeatedly growing a fundamental unit comprising a GaAs (well) and an Al_{0.3}Ga_{0.7}As (barrier) layers. The layers are thin enough to allow for wave-function overlap and formation of energy mini bands.⁴ A natural extension of the periodic problem is the introduction of disorder by either making the well and/or barrier widths or the barrier composition (x) vary in a random fashion. An intentionally disordered superlattice (DSL) that is nonperiodic in the growth direction, is thus generated.^{5,6} A profound consequence of disorder is the localization of electronic states that is known to shrink the effective band gap or the mobility gap. Disordered superlattices have been studied by several researchers before with the purpose of observing localization effects on vertical transport and the density of states.⁷⁻⁹ We observe a red shift in the heavy-hole excitonic PL peak and a much stronger PL in the nonperiodic superlattices as compared to that in the periodic superlattices. Unusual optical properties¹⁰⁻¹³ and electronic structure¹⁴⁻¹⁶ have been predicted and observed for AlAs/GaAs and Si_{1-x}Ge_x Si disordered superlattices. We address here the reasons for the stronger PL in the DSLs. We interpret the stronger PL in the nonperiodic superlattices in terms of an enhanced free-exciton radiative decay rate that is

a result of a breakdown in the translational symmetry. We present the PL data and arguments to support this interpretation.

Let us first compare the exciton in a 3D crystal with that in a quantum well. In a 3D crystal the exciton and photon form a stationary polaritonic state that in principle does not decay.¹⁷⁻¹⁹ Intrinsic radiative (spontaneous) decay is not possible because both energy and momentum cannot be conserved in such a transition.²⁰ The radiative decay observed as PL is a result of interaction of the polariton with the crystal boundaries or due to the symmetry breaking presence of defects and impurities.²¹ Defects and impurities localize or bind the exciton and shorten its lifetime (or increase the radiative decay rate or the oscillator strength). The term "giant oscillator strength" is used in connection with bound excitons and was calculated by Rashba and Gurgenishvili²² and has been used to account for the bound excitonic transitions observed in GaAs at impurity concentrations as low as 10^{-10} cm⁻³. The situation in quantum wells is quite different from that in 3D crystals. The inherent lack of translational symmetry now allows for spontaneous decay of the free exciton. The basic idea is that, in lower-dimensional systems, the breakdown in translational symmetry in one or more directions modifies the coupling of the exciton to photons. In quantum wells, the quasi-2D exciton with an in-plane wave vector of magnitude $|\mathbf{k}_\parallel| < n\omega/c$, where ω is the frequency of the radiation at the exciton energy, n is the refractive index and c is the speed of light, can couple to a range of photons with a distribution in $\mathbf{k}_z$,^{21,23} because of a relaxation of the momentum selection rule. The exciton-photon coupling is therefore very strong in the lower-dimensional systems. Quantum wells thus permit the study of the intrinsic nature of exciton recombination and light-matter interaction.² The calculations of Hanamura²⁴ for excitons in quantum wells, in the 2D limit predict an enhancement of the radiative decay rate by a factor of $24\pi(\lambda/a_b)^2$, where λ is the wavelength corresponding to the exciton energy and a_b is the exciton Bohr radius. Corrections to this factor by including the refractive index and accounting for the quasi-2D nature of the exciton have been considered by Andreani, Tassone, and Bassani.²⁵ The phenomenon of enhanced radiative decay in single GaAs quantum wells was first demonstrated by Deveaud *et al.* using PL.²⁶ An enhanced radiative decay rate implies a larger number of radiative recombinations per unit

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time which in turn implies a stronger PL emission. With regard to the stronger PL in the DSLs we recognize an underlying similarity between the nonperiodic structures and quantum wells in the context of a lack of translational symmetry in the growth direction.

II. EXPERIMENT

Measurements of PL were performed on two sets of GaAs/Al_{0.3}Ga_{0.7}As superlattices each consisting of an ordered or periodic SL (OSL) and a corresponding disordered or nonperiodic SL (DSL) in which the well widths are randomly distributed about a mean value matching that of the OSL. Each sample consisted of 40 GaAs wells separated by Al_{0.3}Ga_{0.7}As barriers of width 9 Å. The period of the OSL with well width $L_w = 77$ Å was 86 Å (86 OSL) and that of the OSL with $L_w = 40$ Å was 49 Å (49 OSL). In the corresponding DSLs the well widths had a mean value of 77 Å with a standard deviation σ of 14 Å in one and a mean of 40 Å with a σ of 12 Å in the other. The samples were grown by MBE on GaAs substrates separated by a 1500 Å buffer of GaAs followed by a layer of Al_{0.3}Ga_{0.7}As of thickness 2000 Å. The measurements were made at 4.2 K and above. The PL was excited using various laser sources and measured by conventional methods using a 0.75 m monochromator followed by detection with a photomultiplier tube and lock-in techniques. The slit width resolution of the spectrometer was 0.5 meV.

The dependence of the PL strength on the excitation wavelength λ_e , excitation power, and temperature were studied. Figure 1(a) shows a comparison of the PL emission spectra between the 49 OSL and DSL, excited at different wavelengths. The excitation power was 50 mW cm⁻². The dominant PL is due to the heavy-hole excitonic transition. The PL from the DSL is eight times stronger for $\lambda_e = 633$ nm and five times stronger for $\lambda_e = 514$ nm. When the excitation wavelength was increased, that is, as we approach the band gap, the ratio, denoted by R , of the PL from the DSL to that in the OSL becomes larger reaching a value of about 20 for $\lambda_e = 750$ nm. The plot in Fig. 1(b) illustrates the dependence of the ratio R on λ_e . In this comparison the excitation power was kept at the same level for both samples. Another observation was that the dominant peak in the DSL is shifted to longer wavelengths from that in the OSL. A red shift of about 30 meV was observed for the 49 DSL and 5 meV for the 86 DSL. A study of this shift was presented in an earlier article.²⁷ The excitation power dependence of the PL peak intensity for the OSL and the DSL are shown in Fig. 2. The abscissa n_{ex} in Fig. 2 is the exciton excitation density which is estimated from the excitation power using $n_{ex} = g\tau_f$, where g is the rate at which carriers are generated by the incident photons and τ_f is the exciton formation time (taken to be 400 ps, which is the value measured for GaAs quantum wells at 10 K).²⁸ The quantity g is given by $\phi\alpha d$ where ϕ is the incident photon flux, α is the absorption coefficient (assumed to be $\sim 10^5$ cm⁻¹ for $\lambda_e = 514$ nm), and d is the sample thickness. The results of the temperature dependence of the PL excited at $\lambda_e = 750$ nm are summarized in Fig. 3. The PL intensity, of the DSL, remains constant between 6

FIG. 1. (a) Photoluminescence spectra at 10 K for the 49 DSL and 49 OSL for two different excitation wavelengths. The excitation power was 50 mW cm⁻². The PL of the OSL has been magnified by the factor shown alongside the spectra. (b) The excitation wavelength dependence of the ratio R of the PL from the DSL to that from the OSL for (●) the 49 DSL and OSL and (■) the 86 DSL and OSL.

and 30 K and decreases for $T > 30$ K, while in the OSL it decreases continuously.

III. DISCUSSION

The differences between the behavior of PL in the OSL and DSL suggest that the PL processes might be different in these two systems. The measured PL emission intensity is proportional to the quantity $n_e\Gamma_{lum}$, where n_e is the density of free excitons and $\Gamma_{lum} (= 1/\tau_{lum})$, where τ_{lum} is the radiative lifetime) is the radiative decay rate. An enhanced radiative decay rate would account for the stronger PL. The total decay rate is inversely proportional to the decay time τ which is given by $1/\tau = 1/\tau_{lum} + 1/\tau_{nr}$ where τ_{nr} is the nonradiative lifetime. Nonradiative decay channels shorten the total decay time τ . However, the observation of an enhanced decay rate, which relies on the coherent nature of the exciton, is impaired by any process that changes the exciton lifetime.²⁶ Only excitons with $k_{||} < n\omega/c$ can decay radiatively.²³ Such excitons are said to belong to radiative states. Any scattering or energy relaxation process such as scattering by acoustical

FIG. 2. Variation of the PL intensity (Y axis on the left-hand side) excited at $\lambda_e = 514 \text{ nm}$ with exciton excitation density for the (■) 49 DSL and (●) 49 OSL. The solid triangles are the ratio of the PL from the DSL to the OSL for the same excitation power and are given by the Y axis on the right-hand side.

phonons or collisions that do not destroy the exciton, but change the $k_{\parallel}$ value of the exciton, will increase τ_{lum} and hence τ . In the model of Deveaud *et al.*²⁶ the long-time solution for τ_{lum} is given by

$$\tau_{lum} = (\tau_0 \tau') / (1/\tau_0 + 1/\tau' + 1/\tau_{deph}).$$

Deveaud *et al.* assume a two-level system of a radiative and a nonradiative states in modeling the exciton decay. The intrinsic lifetime of the radiative state is τ_0 and the exciton is assumed to scatter to the nonradiative state with a dephasing time τ_{deph} and be recaptured to the radiative state with a characteristic time τ' and the above equation is derived based on rate equations. Any process that decreases τ_{deph} will increase τ_{lum} and causes an additional homogeneous broadening. Deveaud *et al.* observe just such a behavior, namely an increase in the homogeneous broadening (decreasing dephasing time) together with an increasing decay time as the excitation density is increased. The behavior is attributed to

FIG. 3. Variation of the PL intensity, excited at $\lambda_e = 750 \text{ nm}$, with temperature for the (■) 49 DSL and (●) 49 OSL.

increased scattering events. Thus, the study of intrinsic exciton decay τ_0 has been elusive. In our experiments we are comparing the PL between two cases where the only difference is the translational symmetry. In the DSLs, the increased number of radiative channels (all states with $k_{\parallel} < n\omega/c$) provided by the breaking down of the translational symmetry results in a shorter radiative lifetime. In such a case the radiative decay time may be expected to dominate the PL, and thus yield a stronger PL emission. Additional localization due to in-plane roughness causes a mixing with the states with higher value of $k_{\parallel}$ (nonradiative states) and the radiative lifetime will increase. In the OSLs, as in a 3D crystal, the free-excitonic PL is due to the radiative decay of the exciton that is localized because of defects such as interface roughness in the case of the OSLs. Excitons localized by interface defects have a longer lifetime than excitons localized purely by a lack of translational symmetry. For a quantum-well structure free of defects, the intrinsic decay times are predicted to be of the order of 10 ps.^{25,29} Localization due to defects increases this time. Calculations show that in a 100 Å GaAs/Al_xGa_{1-x} quantum well the lifetime for the L and T modes is 25.5 ps due to the phenomenon of lack of translational invariance and about 100 ps to 1 ns for excitons localized by defects.^{21,25,26} For this reason, to observe the enhanced decay in quantum wells, brought about purely by the destruction of translational invariance, one needs single quantum wells of high purity. From their measurements in GaAs quantum wells Deveaud *et al.*²⁶ have deduced a radiative lifetime of $10 \pm 4 \text{ ps}$. The measured lifetimes usually are longer because of energy relaxation processes that increase τ_{lum} or because of the influence of in-plane localization. We observe that the PL in the DSLs is always stronger than that in the OSLs. However, our experiments show that the ratio R depends on the conditions of the excitation process. Excitation at energies above the band gap creates initially a nonthermal distribution of excitons which then relax. The higher energy of the excited carriers and the slow cooling rate of the carriers cause an increase in τ_{lum} and consequently reduces the decay rate.³⁰ The effects of the intrinsic nature of the exciton in general become more obvious for excitation energies nearer the band gap. In our samples this manifests as an increase in the ratio R with increase in λ_e . The same reasoning accounts for the bound exciton transitions observed in GaAs even at low-impurity concentrations for band-gap excitations,²² the near-band-gap PL from a GaAs/AlGaAs interface,³⁰ and also for the increase in the free-exciton lifetime in GaAs when the excitation photon energies are increased above the band gap of GaAs.²⁹

At higher excitation powers the increase in exciton-exciton scattering events decreases τ_{deph} and increases τ_{lum} . The radiative decay time is then dominated by these events which preclude the observation of the intrinsic behavior and the ratio R decreases. The effects of τ_{deph} on the power dependence of the PL in the DSL is different from that in the OSL because τ_0 is different. In the same power range, with increasing power, the PL intensity increases with different slopes for the OSL and for the DSL with an effective decrease in the ratio R . The difference in lifetimes is expected to lead to differences in saturation behaviors of the OSL and

DSL. We do not think that saturation is observed in either sample in the power range studied.

We consider next the constancy of the PL intensity with temperature (Fig. 3) in the temperature range of 6–30 K, in the DSL. The density of free excitons n_e in radiative states ($k_{\parallel} < n\omega/c$) increases linearly with temperature while the decay rate Γ_{lum} decreases with temperature and is proportional to $1/T$.^{21,23} This could account for the product $n_e \Gamma_{\text{lum}}$, which gives the PL intensity, being constant with temperature.

From the discussion above it is clear that while the ratio R can be an indication of enhanced radiative decay, it cannot provide any quantitative measure because of its sensitivity to the excitation conditions and the sample quality. A true value of R can be obtained only for structures completely free of defects. Strong PL intensities have been reported for disordered AlAs/GaAs and AlP/GaP superlattices.^{10–12,31–33} The corresponding ordered superlattices have an indirect band structure. Mäder and co-workers have performed pseudopotential calculations with plane-wave basis and show that for the disordered AlAs/GaAs superlattices the band gap becomes direct in the planar Brillouin zone.¹⁶ This leads to a greater oscillator strength. A further enhancement occurs because of a relaxation of $k_{\parallel}$ selection rules. Both these reasons contribute to the much larger value of R observed in disordered AlAs/GaAs and possibly in the AlP/GaP superlattices. Short PL lifetimes measured by time-resolved PL have been reported by Kasu *et al.* for disordered AlAs/GaAs superlattices.³⁴ For the composition of our samples the band gap remains direct in the DSL. The enhancement of the PL in our DSLs is therefore attributed to the disorder-induced presence of additional radiative channels made possible by a relaxation of $k_{\parallel}$ selection rules. Wakahara *et al.*¹³ have argued, however, that in the case of disordered SiGe strained layers, carrier confinement rather than localization effects are responsible for the enhanced PL, based on their observation that the PL behavior in the SiGe DSL is similar to that in the corresponding OSL. In conclusion, the observed differences in the PL behavior between the ordered and disordered GaAs/Al_{0.3}Ga_{0.7}As superlattices, studied in this work, are explained by invoking the rapid radiative decay of free excitons brought about by a breakdown in translational symmetry in superlattices with deliberately introduced nonperiodicity. The enhanced intrinsic radiative decay rate is best observed at low temperature, low power, and resonant excitation in high-quality samples. Quantitative estimates need time-resolved spectroscopy.

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