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Direct coupling of heavy-hole free excitons in $In_xGa_{1-x}As/GaAs$ quantum wells with free excitons in the GaAs barrier

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Enhanced emission due to heavy-hole free-exciton (HHFE) collapse in narrow ($L_z \leq 50$ Å) single $In_{0.1}Ga_{0.9}As/GaAs$ quantum wells was observed when the excitation energy was resonant with the GaAs-barrier free-exciton energy. The observed resonant excitation behavior along with the marked difference in the effect of an applied magnetic field on the HHFE emission strength for the cases of nonresonant and resonant excitation is consistent with a model of direct coupling between the GaAs-barrier free-exciton level and the $In_{0.1}Ga_{0.9}As$-quantum-well heavy-hole free-exciton level. The direct coupling between the GaAs-barrier free-exciton level and the quantum-well HHFE level is a result of wave-function overlap accomplished by extension of the HHFE wave function into the barrier region. As a result of this direct coupling, when several $In_{0.1}Ga_{0.9}As$ quantum wells of different sizes are present in the same sample, excitation at the GaAs-barrier free-exciton formation energy simultaneously resonantly excites HHFE emission from all of the $In_{0.1}Ga_{0.9}As$ wells for the well sizes studied ($L_z \leq 50$ Å).

The low-temperature photoluminescence (PL) spectrum of a high-quality, undoped $Al_xGa_{1-x}As/GaAs$ quantum-well structure is usually dominated by emission due to collapse of heavy-hole free excitons (HHFEs) located in the well region. Emission from the barrier region is largely suppressed, even if the excitation energy lies at or above the barrier band gap, since carrier transfer from the barrier into the well is very efficient. When the excitation energy is resonant with a quantum-well transition which lies above the HHFE in energy, efficient transfer between the laser-excited state and the HHFE may result, and enhanced HHFE emission may be observed. Kusano *et al.*[1] showed that laser excitation of the light-hole free exciton (LHFE) leads to enhanced emission due to HHFE collapse. Using time-resolved luminescence techniques with picosecond resolution, they showed that the kinetics governing the formation of HHFEs from LHFEs differ significantly from those governing the formation of HHFEs from free electrons and holes. Since the wave-function overlap between the initial state (LHFE) and final state (HHFE) is great, the LHFE-to-HHFE transition is a favorable process. Energy conservation is accommodated by phonon emission.

In quantum-well structures for which the barrier is a binary compound, for example, the $In_xGa_{1-x}As/GaAs$ system, an additional resonant excitation pathway arises: direct excitation of excitons in the barrier may lead to enhanced emission from the quantum well. In this paper we examine the luminescence properties of pseudomorphically strained, thin ($L_z \leq 50$ Å) $In_{0.1}Ga_{0.9}As$ single quantum wells (SQWs) with GaAs barriers grown on GaAs substrates. We find that HHFE emission from the $In_{0.1}Ga_{0.9}As$ wells is greatly enhanced when the excitation energy is tuned to the energy for free-exciton formation in the GaAs barrier. We find that this effect is independent of well size for all samples investigated ($L_z \leq 50$ Å). Additionally, we examine the effect of an applied magnetic field on the luminescence properties of the SQW structures under both resonant and nonresonant excitation conditions.

The single quantum-well structures studied consisted of a single 2-, 4-, 6-, 10-, or 17-monolayer (ML) -thick (1

ML = 2.85 Å) $In_{0.1}Ga_{0.9}As$ quantum well sandwiched between 500-nm GaAs barriers, with the buffer layer being simply the first GaAs barrier. The structures were grown on n^+-type GaAs(001) substrates by conventional solid source molecular-beam epitaxy (MBE) in a Varian Gen II MBE machine using a tetrameric arsenic source. The GaAs and $In_{0.1}Ga_{0.9}As$ growth rates were 0.90 and 1.00 ML/sec, respectively. The GaAs and $In_{0.1}Ga_{0.9}As$ growth temperatures were 580 and 540 °C, respectively, except the last 35 nm of the first barrier, for which the substrate temperature was ramped from 580 to 540 °C. Growth interruption was not used.

The photoluminescence (PL) was excited with a tunable dye laser using Styryl 9 dye, which was pumped with an Ar^+-ion laser. The pump power used in all experiments in this study was approximately 500 mW/cm^2. The PL measurements were made at 2 K with the sample immersed in liquid He. A magnetic field oriented perpendicular to the growth direction was used to observe the diamagnetic shifts of the PL transitions, as well as to observe the PL intensity variation with magnetic field for both resonant and nonresonant excitation. The PL spectra were analyzed with a high-resolution 4-m spectrometer equipped with a RCA C31034A photomultiplier tube for detection.

The PL spectra observed for each sample showed two main features: the normal GaAs band-edge emission peaks and the $In_{0.1}Ga_{0.9}As$ quantum HHFE emission peak. When a magnetic field was applied normal to the growth direction, the HHFE transition energy showed the expected blueshift. In Fig. 1 the observed diamagnetic shift of the quantum-well HHFE transition for an applied magnetic field of 36 kG is shown as a function of well size for well sizes between 6 and 50 Å (between 2 and 17 ML). The diamagnetic shift is seen to increase roughly linearly with decreasing well size. The solid line connecting the data points extrapolates at zero well width to 1.2 meV, which is the expected value[2] for the diamagnetic shift of bulk GaAs for the same applied magnetic-field strength. This is understood on the basis that as the well size decreases the HHFE wave function penetrates further into the GaAs barrier and thus the HHFE takes on more and more GaAs character. It is worth noting that for the $Al_{0.3}Ga_{0.7}As$/GaAs system the diamagnetic shift observed for the GaAs quantum-well HHFE transition decreases as the well size decreases, which is opposite to the trend observed here. The penetration of the quantum-well exciton wave function into the barrier was previously demonstrated in the $In_{0.1}Ga_{0.9}As$/GaAs system by Kirby, Constable, and Smith.[3] They found that the HHFE linewidth decreased as the well width decreased and concluded that the linewidth was primarily influenced by alloy broadening in the $In_{0.1}Ga_{0.9}As$ well. As the well size decreases more of the wave function extends into the barrier, which in this case is a binary, thus reducing the alloy scattering and thereby resulting in narrower lines. We have observed the same trend: the HHFE PL linewidths (full width at half maximum) for the 6-, 4-, and 2-ML quantum wells were 0.54, 0.22, and 0.11 meV, respectively. The different structures studied differed only in the width of the quantum well, and the results of our observations were very similar for each of these structures. For this reason, below we show results for only the 6-ML quantum-well sample, while it is understood that similar results were obtained for the other SQW structures.

Figure 2 shows the selective excitation spectra obtained for the 6-ML SQW structure with the detector hold position at the HHFE energy of 1.5093 eV. The peak at 1.5190 eV is the GaAs band edge, and the peak at 1.5150 eV is the GaAs free-exciton creation energy. The large increase in intensity in the lowest-energy region is due to the increase in scattered laser light which occurs when the excitation energy approaches that of the detector hold position. Both the GaAs band edge and the GaAs exciton peaks are seen to resonantly excite the $In_{0.1}Ga_{0.9}As$ well HHFE transition; however, excitation at the GaAs exciton energy is significantly more efficient in producing subsequent HHFE emission.

In Fig. 3 we show the resonant excitation spectra of the 6-ML SQW structure for various excitation energies. The data show strong emission due to HHFE collapse when

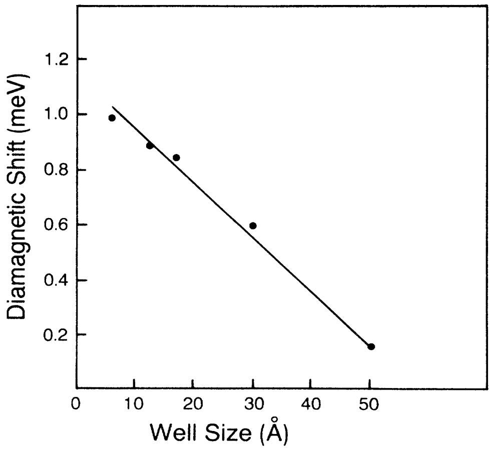

FIG. 1. Diamagnetic shift of the HHFE transition as a function of well size for five different $In_{0.1}Ga_{0.9}As$/GaAs single quantum-well structures.

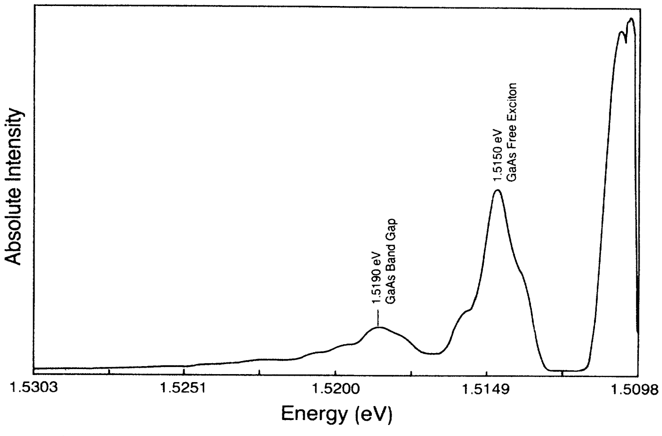

FIG. 2. Selective excitation for the 6-ML $In_{0.1}Ga_{0.9}As$/GaAs single quantum-well structure. The detector hold position is the HHFE energy of 1.5093 eV.

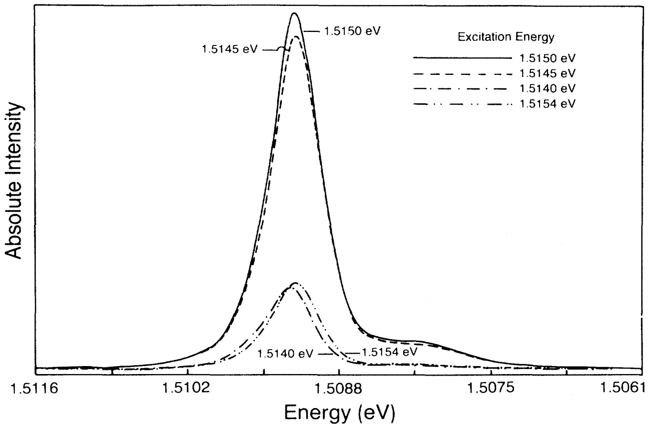

FIG. 3. Resonant excitation spectra of the 6-ML $In_{0.1}Ga_{0.9}As$/GaAs single quantum-well structure obtained using direct excitation of the GaAs-barrier free exciton at 1.5150 eV.

the excitation energy is resonant with the GaAs exciton formation energy, while excitation energies just a few tenths of a meV both above and below the GaAs free-exciton energy results in considerably less HHFE emission, demonstrating the resonant nature of the excitation. Since free carriers are not produced at the GaAs free-exciton formation energy, the resonant behavior observed is due to a transition involving the GaAs-barrier free-exciton and the quantum-well HHFE. If the HHFE formation process from the GaAs free exciton involved recombination of the GaAs free exciton followed by absorption of the emitted photon to create free carriers in the well, followed by subsequent HHFE formation, then the overall (GaAs free exciton→ HHFE) process would be expected to be very inefficient, in contrast to our observations. It is most likely that the GaAs free exciton is directly coupled to the quantum-well HHFE level.

Resonant excitation spectra for the 6-ML SQW structure are shown in Fig. 4 in the HHFE emission region for different applied magnetic-field strengths. The excitation energy is resonant with the GaAs free-exciton formation energy, 1.5150 eV. Strong emission is observed at zero applied magnetic field, while increased applied magnetic fields lead to a dramatic decrease in HHFE emission, in contrast to usual observations regarding excitonic transition strengths in magnetic fields. The observed behavior is understood on the basis of the proposed, direct (GaAs-barrier free exciton→ HHFE) process as follows: The magnetic field compresses the wave functions of both the GaAs-barrier free exciton and the $In_{0.1}Ga_{0.9}As$ well HHFE, leading to a reduced coupling between the two exciton levels due to the reduction in wave-function overlap. Thus the observed magnetic-field-dependent resonant excitation behavior of the HHFE transition is consistent with the proposed mechanism of direct HHFE formation from the GaAs-barrier exciton.

The HHFE emission strength was observed to decrease significantly with increasing applied magnetic field under resonant excitation conditions, as just discussed and shown in Fig. 4. In marked contrast, under nonresonant excitation conditions the HHFE emission strength is found to increase with increasing applied magnetic-field strength. Figure 5 shows the variation of HHFE emission strength as a function of applied magnetic-field strength using a nonresonant excitation energy of 1.5253 eV, which is 6.3 meV above the GaAs-barrier band gap. In this case free electrons and holes are created which migrate to the well and generate the HHFE in the well. The presence of an applied magnetic field probably does not markedly affect the rate at which the free carriers migrate into the well region and form HHFEs. However, once the HHFE is formed, its recombination rate is enhanced because its wave function is compressed by the applied magnetic field. The observed differences in magnetic-field-dependent behavior between the cases of resonant and nonresonant excitation is consistent with the model of direct coupling of the GaAs-barrier free exciton with the quantum-well HHFE (when resonant excitation is used).

We have also found that when several $In_{0.1}Ga_{0.9}As$ quantum wells of different sizes ($L_z \leq 50$ Å) are grown in

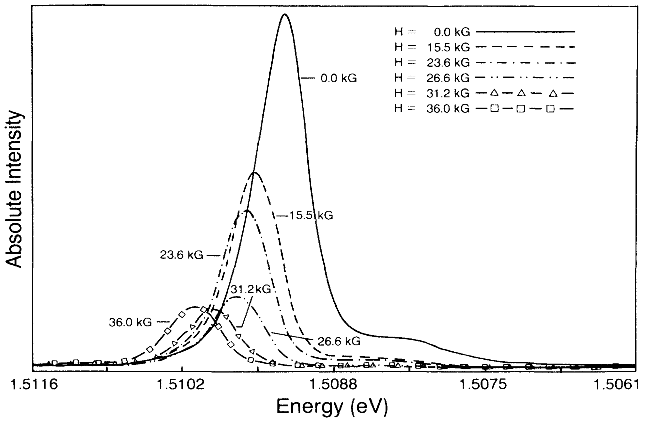

FIG. 4. Resonant excitation of the 6-ML $In_{0.1}Ga_{0.9}As$/GaAs single quantum-well structure showing the intensity variation of the HHFE transition as a function of applied magnetic-field strength.

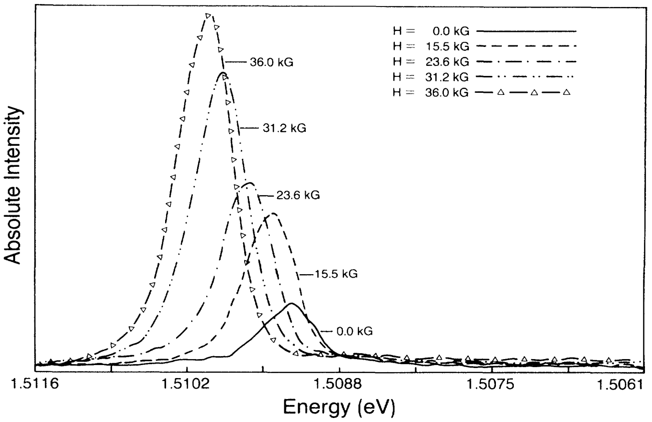

FIG. 5. Nonresonant excitation of the 6-ML $In_{0.1}Ga_{0.9}As$/GaAs single quantum-well structure showing the intensity variation of the HHFE transition as a function of applied magnetic-field strength.

the same sample, resonant excitation of the GaAs free exciton leads to simultaneous resonant excitation of all of the $In_{0.1}Ga_{0.9}As$ QWs. The situation is different for the $Al_{0.3}Ga_{0.7}As/GaAs$ system for which each quantum-well size requires a different resonant excitation energy.

The GaAs-barrier free exciton is coupled to the quantum-well HHFE via the wave-function penetration of the HHFE into the barrier region. Such a coupling also must satisfy energy and momentum conservation rules. Since the total angular momentum of holes associated with both the GaAs barrier free exciton and the quantum-well HHFE states is $J=\frac{3}{2}$, total angular momentum conservation is ensured.

The situation is somewhat similar for coupling between the LHFE and HHFE levels in $Al_{0.3}Ga_{0.7}As/GaAs$ quantum wells. The wave-function overlap is obviously great because the LHFE and HHFE occupy the same region in the quantum well and do not differ markedly in size. However, the light- and heavy-hole levels differ in m_J value ($m_J=\pm\frac{1}{2}$ and $m_J=\pm\frac{3}{2}$, for light and heavy holes, respectively). Since the hole associated with the GaAs-barrier free exciton has mostly heavy-hole character,[4] a change in m_J value is not required for the (GaAs free exciton$\rightarrow$ HHFE) process studied here for the $In_{0.1}Ga_{0.9}As/GaAs$ system.

In summary, enhanced emission due to HHFE collapse in single $In_{0.1}Ga_{0.9}As$ quantum wells was observed when the excitation energy was resonant with the GaAs-barrier free-exciton energy. The observed resonant excitation behavior along with the marked difference in the effect of an applied magnetic field on the HHFE emission strength for the cases of nonresonant and resonant excitation is consistent with a model of direct coupling between the GaAs-barrier free-exciton level and the $In_{0.1}Ga_{0.9}As$ quantum-well heavy-hole free-exciton level. As a result of this direct coupling, when several $In_{0.1}Ga_{0.9}As$ quantum wells of different size are present in the same sample, excitation at the GaAs-barrier free-exciton formation energy simultaneously resonantly excites HHFE emission from all of the $In_{0.1}Ga_{0.9}As$ wells for the well sizes studied ($L_z \leq 50$ Å).

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