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Radiative Recombination At the Alxga1-Xas-GaAs Heterostructure Interface by 2-Dimensional Excitons

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Radiative recombination from the $Al_xGa_{1-x}As$-GaAs heterostructure interface was investigated using photoluminescence and photoluminescence-excitation spectroscopy in modulation-doped and undoped samples. This emission is identified as H-band *A*, resulting from an indirect excitonlike transition in real space. The exciton is made up of a two-dimensional electron in the interface notch and a valence-band hole in the neutral region, having a binding energy of 1.8 meV. The H-band *A* exciton is directly excited by a free exciton making a vertical transition in real space. H-band *A* may be a distortion of the vertical direct free exciton in real space to an indirect free exciton in real space, in which case only one exciton is involved; or it could result from the direct excitation of the indirect exciton in real space by the vertical exciton through wave-function overlap, with energy and momentum being conserved. In the latter case two excitons are involved. While the exact coupling mechanism between the free exciton and the H-band *A* exciton is not well understood, two possibilities are suggested.

INTRODUCTION

Since the advent of modulation doping in semiconductor structures[1,2] a great deal of effort has gone into understanding the characteristic band structure of the $Al_xGa_{1-x}As$–GaAs single heterostructure. Much of the effort has been focused on the radiative-recombination processes associated with the heterojunction interface. The unique characteristics of this radiative recombination were initially reported by Yuan *et al.*[3,4] The emission consisted of a broad line occurring on the low-energy side of the bulk GaAs bound-exciton emission. This broad emission line was identified as the H band. The spectral shape and position of this band was strongly sample dependent as well as dependent on temperature and excitation intensity and energy. The source of the H-band emission was attributed to recombination between the two-dimensional electron gas (2DEG) and free holes in the GaAs neutral region, well separated from the 2DEG. The 2D electrons are contained in the notch potential at the heterostructure interface. Yuan *et al.* first attributed the H-band emission to 2D electrons that tunnel away from the notch potential at the interface and recombine with free holes in the flat-band region. The H-band emission was later interpreted by Balsley[5] as resulting from weakly bound interface excitons, in analogy with excitons in quantum wells in an external electric field.

Two different H-band recombination channels were observed by Kukushkin, Klitzing, and Ploog,[6] which they labeled *A* and *B*; band *A* resulted from recombination of 2D electrons with free holes in the valence band, while band *B* resulted from the recombination of 2D electrons with holes bound to acceptors. Both H bands shift to higher energy with increasing excitation intensity. A similar behavior was observed by Zhao *et al.*[7] when the modulation-doping concentration was increased. The intensity dependence results from a change in the potential across the GaAs active layer; i.e., increased excitation intensity results in neutralizing ionized acceptors, which reduces the band bending with a resultant increase in the H-band emission energy. With decreased band bending a decrease in the linewidth of the H band is also observed.

It has been generally believed that H-band *A* results from the recombination of 2D electrons with photoexcited free holes.[8–12] It is the purpose of this paper to instead show that H-band *A* itself has an excitonic nature and, moreover, is directly excited by a free exciton. The exact nature of the free exciton is still an open question; it may be a 2D exciton, defined as an exciton formed from a 2D electron, associated with the 2DEG, and a valence-band hole in a vertical transition in real space; or it could be the bulk GaAs free exciton, also resulting from a vertical transition. We believe that H-band *A* (HB_A) results from an excitonlike transition involving a 2D electron, associated with the 2DEG, and a valence-band hole in an indirect transition in real space. The direct excitation of HB_A may result from a simple one-step process in which the hole of an existing 2D exciton is moved away from the interface notch by the space-charge potential, while the 2D electron remains in the notch. The H-band exciton would then recombine in an indirect transition in real space. An alternative direct one-step excitation could begin with the GaAs free exciton located in the GaAs flat-

band active layer. The electron of this exciton could be drawn towards the interface notch by the space-charge potential, while the hole would remain in the active layer. The exciton then would recombine in an indirect transition in real space, exhibiting the lower HB_A emission energy. On the other hand, the excitation may result from direct coupling of the vertical free exciton in real space with the nonvertical HB_A exciton. Such a direct coupling would result from wave-function overlap of the free exciton with the HB_A exciton, analogous to the direct coupling of the GaAs exciton in the barrier with the $In_xGa_{1-x}As$ exciton in the well in $In_xGa_{1-x}As$-GaAs quantum wells.[13] In this case two independent excitons are involved as opposed to the case above, where the direct free exciton realigns into the indirect HB_A exciton. The exact coupling mechanism is still to be resolved. We also should consider the possibility that the resonance at the exciton energy might result from an increased absorption coefficient at the exciton energy, with the exciton then breaking up in the notch field, providing free carriers. H-band *A* would then proceed from recombination of a 2D electron and a free hole. Experimental data will show that this is not the correct explanation.

Two different samples were investigated. One was a nominally undoped sample in which the notch region at the interface between $Al_xGa_{1-x}As$ and GaAs was formed from residual impurities. In this sample the GaAs active region was 0.5 μm wide. The other sample also had a single heterojunction, but differed in that the $Al_xGa_{1-x}As$ barrier was heavily modulation doped and the GaAs active region was 500 Å wide. A 500-Å active region results in a small but measurable confinement. Very similar excitation of HB_A was observed in both samples, i.e., the photoluminescence-excitation (PLE) spectrum showed a sharp resonance at the 2D exciton energy.

EXPERIMENTAL PROCEDURE

The modulation-doped sample studied was an $Al_xGa_{1-x}As$-GaAs heterostructure grown by molecular-beam epitaxy (MBE) on a semi-insulating GaAs substrate, using a Varian MBE machine. The substrate was nominally oriented 6° from (100) towards the (111) Ga plane. An Epi Chorus valved cracker cell was used to produce dimeric arsenic as the arsenic growth species. The GaAs and $Al_xGa_{1-x}As$ growth rates were 0.72 and 1.0 ML/sec (1 ML = 2.8275 Å), respectively, and the As_2 to Ga (uncorrected) beam-pressure ratio was 18, which gave an As-stabilized (2×4) surface reconstruction during GaAs growth at 600±10 °C. The buffer-layer sequence consisted of 500-Å GaAs followed by a ten-cycle [(30-Å $Al_xGa_{1-x}As$)/(30-Å GaAs)] superlattice, followed by 500-Å GaAs, followed by 20-Å undoped $Al_xGa_{1-x}As$, followed by 400-Å uniformly doped (2.5×10^{18}/cm^3 Si) $Al_xGa_{1-x}As$ with an x value of 0.28, as determined by reflection high-energy electron-diffraction oscillations. The structure was terminated with a 350-Å GaAs cap doped with 4×10^{18}/cm^3 Si. In the nominally undoped sample the notch region at the interface between $Al_xGa_{1-x}As$ and GaAs was formed from the presence of residual impurities. This sample was grown by MBE using desorption mass spectrometry from real-time control of $Al_xGa_{1-x}As$ composition.[14] The photoluminescence (PL) and PLE spectra were excited with a tunable dye laser using Styrl 9 dye, which was pumped with an Ar^+-ion laser. The maximum pump power used was approximately 500 mW/cm^2. All measurements were made at 2 K with the sample immersed in liquid He. The spectra were analyzed with a high-resolution 4-m spectrometer equipped with an RCA31034A photomultiplier tube for detection.

EXPERIMENTAL RESULTS

The emission intensity of HB_A as a function of pump energy for the undoped sample is shown in Fig. 1. Also shown is the bound exciton (*B-X*) region and the free-to-bound (*F-B*), free-hole-to-bound-electron, and the bound-to-bound (*B-B*), bound-electron to bound-hole transitions. It is noted that as the pump energy (see legend in Fig. 1) approaches the free-exciton energy, a strong resonance is observed in the intensity of HB_A. It might at first be thought that this resonance is related to the peak in the absorption coefficient at the exciton energy in GaAs, as reported by Sturge.[15] However, in Struge's work, the magnitude of the absorption coefficient at the exciton energy is less than 20% greater than its minimum value on the high-energy side of the free exciton. The lowest pump energy in Fig. 1 is still a little more than 1 meV higher than the exciton energy, so the intensity of HB_A will increase even more as the pump en-

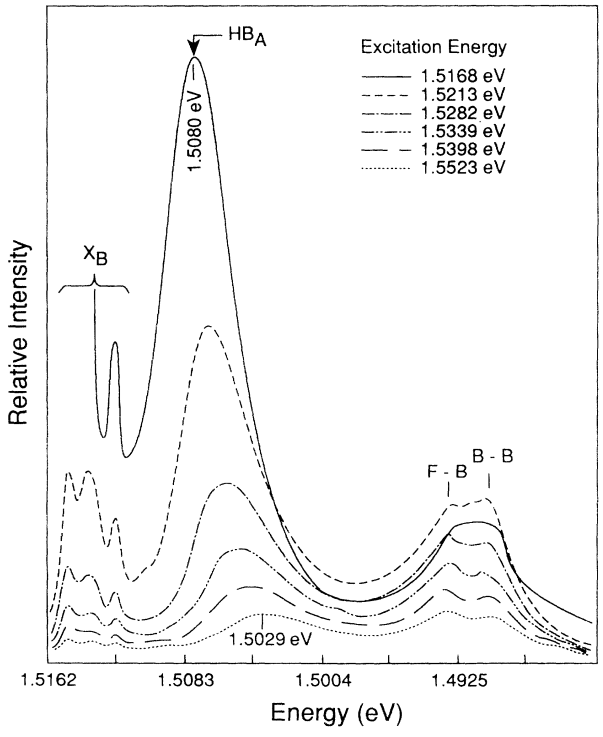

FIG. 1. Emission intensity of the undoped sample as a function of pump energy for X_B, HB_A, *F-B*, and *B-B* transitions. The pump energies are shown in the inset.

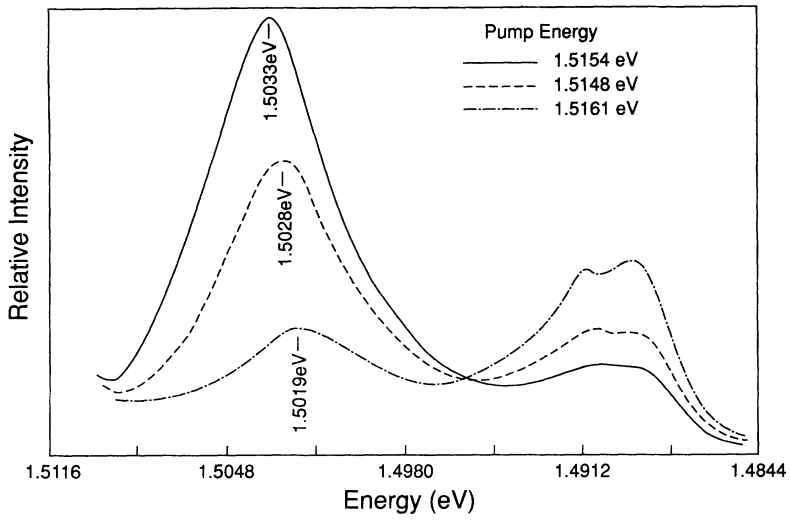

FIG. 2. PL spectra from a companion sample to the one shown in Fig. 1. The intensities of HB_A, *F-B*, and *B-B* are shown at a pump energy resonant with the free-exciton energy and at pump energies slightly above and below the resonant energy.

ergy becomes coincident with the exciton energy. Therefore, the increase in intensity of HB_A cannot be accounted for by the increase in the absorption coefficient at the excitation energy, as the total observed increased is much greater than 20%. It is further noted that the intensity of the *F-B* and *B-B* transitions begins to decrease as the pump energy approaches resonance. If the excitons were breaking up and providing free electrons and holes, one would not expect the *F-B* and *B-B* transitions to decrease in intensity. It might be argued that the impurity levels are becoming saturated; however, it is noted that the bound-exciton transitions are increasing in intensity in nearly the same ratio as H-band *A*, which contradicts this argument. This shows that near resonance, the exciton transitions dominate and that HB_A is resonantly pumped by the free exciton.

In Fig. 2, PL spectra from a second piece of the undoped sample are shown with the pump-beam energy directly on the exciton resonance. When the pump-beam energy is moved a fraction of a millivolt above or below resonance, the intensity of HB_A drops dramatically. The change in absorption coefficient for this small change in pump energy is trivial, contradicting the suggestion that the resonance can be attributed to the peak in absorption coefficient at the exciton energy. It is further noted that the *F-B* and *B-B* transition intensities are at a minimum at resonance, which also contradicts the idea that the excitons are providing free electrons and holes. The emission energy of HB_A is maximum directly at resonance; moving off resonance, either higher or lower, shifts the emission peak to a lower energy. This is expected since the maximum intensity occurs at resonance, which in turn produces the lowest band bending. Moving off resonance in either direction will reduce the intensity, resulting in increased band bending, thus moving the emission peak to lower energy. These results are consistent with the direct excitation of HB_A by the free exciton.

The results of a PLE experiment with the detector-hold position at H-band *A* are shown in Fig. 3. The dominant contribution to HB_A is the free exciton. In this sample the free exciton has essentially the same energy as the bulk GaAs exciton.

If one moves the pump-beam energy off resonance, the emission energy as well as the linewidth of HB_A can still be shifted by changing the pump intensity. This is shown in Fig. 4 where the pump-beam energy is 1.5214 eV. The

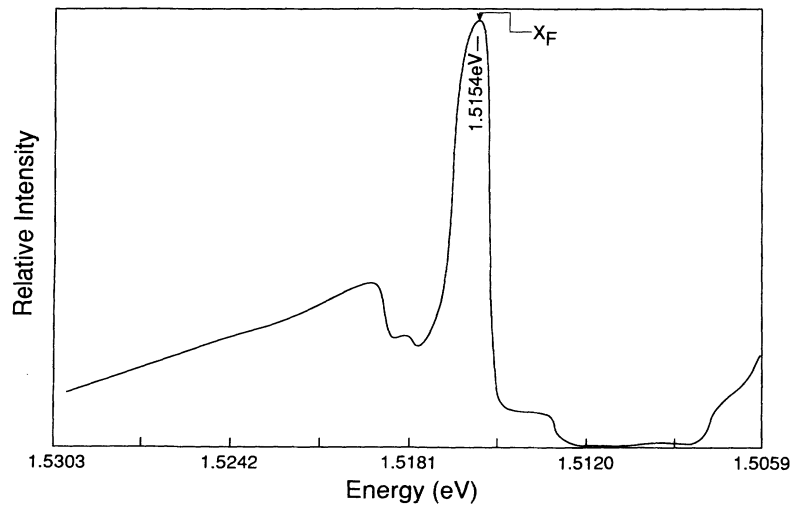

FIG. 3. PLE spectra from the sample shown in Fig. 1, with the detector-hold position at HB_A.

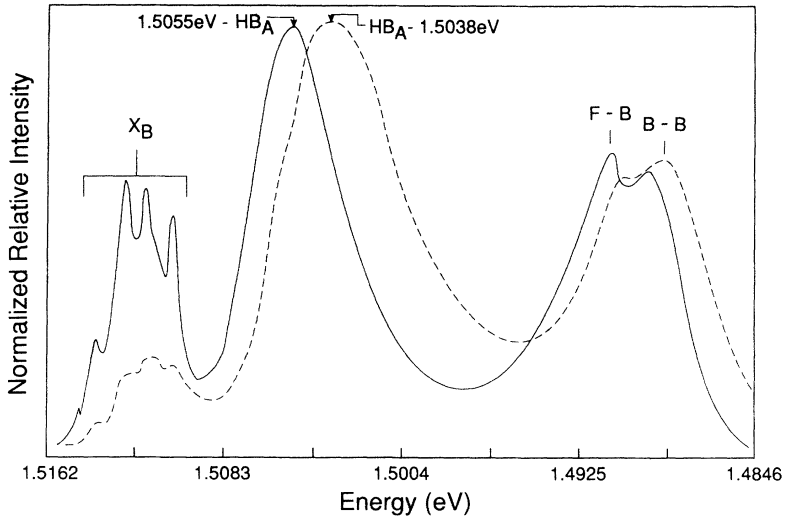

FIG. 4. The shift in emission energy as well as the change in linewidth of HB_A as the intensity of the excitation beam is changed.

pump-beam intensity for the solid curve is a factor of 6 greater than that for the dashed curve. The HB_A emission intensity is normalized to one for both curves to better show the shift in emission energy as well as the decrease in linewidth at the higher pump-beam intensity. The intensities of the different emission features are relative for both curves. Here again as the pump intensity is increased, the emission peak for HB_A moves to higher energy. The greater pump-beam intensity reduces the band bending, shifting the emission to higher energy. The linewidth will also be reduced at higher pump energy since the energy spread of the valence-band holes contributing to HB_A will be reduced because of reduced band bending.

The PL spectra for the doped sample at three different exciting energies are shown in Fig. 5. In this sample the resonant energy of the free exciton is 1.5190 eV, slightly greater than for the undoped sample. This is primarily due to confinement since the sample has a 500-Å GaAs active layer. Here, as was the case with the undoped sample, the intensity of the *F-B* and the *B-B* transitions relative to H-band *A* at resonance is reduced compared to that of the same transitions when slightly off resonance. This is consistent with the conclusion that the free exciton is not providing free electrons and holes. This sample also shows that the impurity transitions are not being saturated since the bound-exciton transitions increase in intensity at a rate similar to that of H-band *A*. These results are consistent with the direct excitation of HB_A by the free exciton. This sample also shows a sharp resonance. The intensity of HB_A falls dramatically when the pump energy is increased or decreased slightly from the resonant energy. Again the H-band *A* emission energy shifts to lower energy as one moves off resonance in either direction.

The PLE spectra for the doped sample are shown in Fig. 6. As in the case of the undoped sample, the detector-hold position is H-band *A*. The dominant contribution to the emission of HB_A is the free exciton, which is consistent with the direct excitation of HB_A by the free exciton. The coupling of the free exciton to HB_A via wave-function overlap must satisfy energy- and momentum-conservation rules. The exciton consists of a

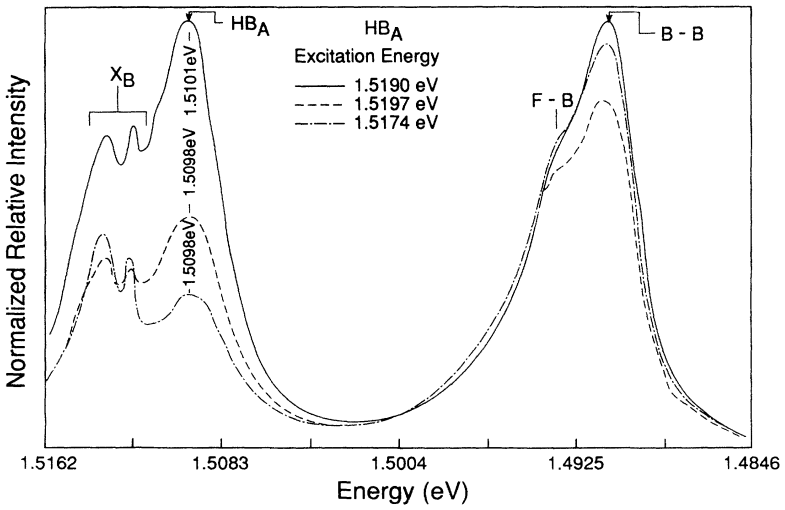

FIG. 5. Emission intensity of the doped sample as a function of pump energy for X_B, HB_A, *F-B*, and *B-B* transitions. The pump energies are shown in the inset.

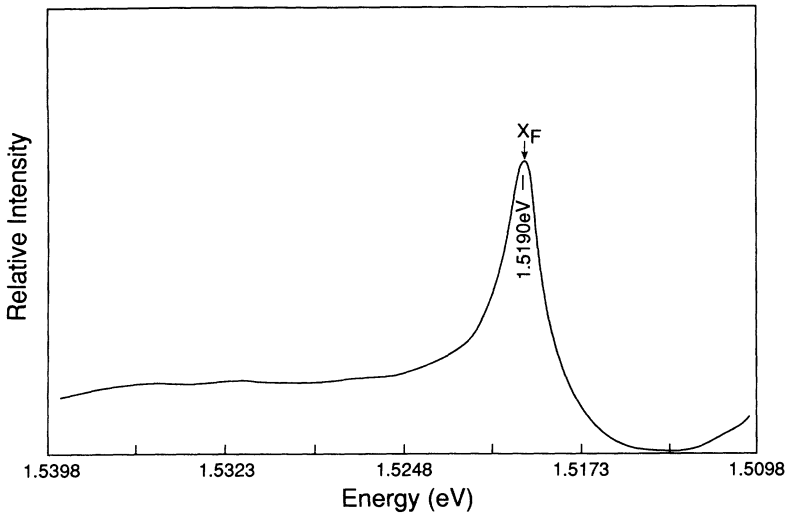

FIG. 6. PLE spectra for the sample shown in Fig. 5, with the detector-hold position at HB_A.

$J=\frac{1}{2}$ electron and a $J=\frac{3}{2}$ hole as does HB_A, so that conservation of total angular momentum would be automatically satisfied. Energy conservation would occur through phonon emission.

DISCUSSION

The maximum in HB_A intensity at the free-exciton pump energy can be accounted for by either of two similar one-step mechanisms. In both mechanisms, HB_A results from an indirect recombination in real space of a free exciton distorted by the space-charge potential, whose electron resides in the interface notch and whose hole resides in the GaAs neutral region. Thus, the maximum number of free excitons at resonance produces a maximum in HB_A intensity.

The first mechanism is shown schematically by the solid lines in Fig. 7. The schematic applies specifically to the doped sample with a 500-Å GaAs active layer. The vertical transition in real space is the 2D exciton, and the indirect transition in real space in the HB_A exciton. The hole of the 2D exciton is moved away from the interface notch by the space-charge potential, while the 2D electron remains in the notch. The exciton then recombines in an indirect transition in real space as the HB_A exciton is reduced in energy due to band bending.

The second mechanism is shown schematically by the dashed lines in Fig. 7. The free exciton in this case is the GaAs exciton in the flat-band region of the active layer. The electron from this exciton is drawn towards the interface notch by the space-charge potential, while the

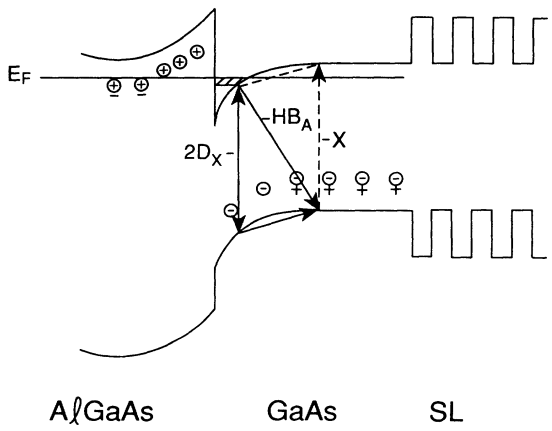

FIG. 7. Energy-level diagram of a single modulation-doped heterostructure. The solid lines show the excitation mechanism of HB_A by the 2D exciton. The dashed lines show the excitation mechanism of HB_A by the GaAs free exciton.

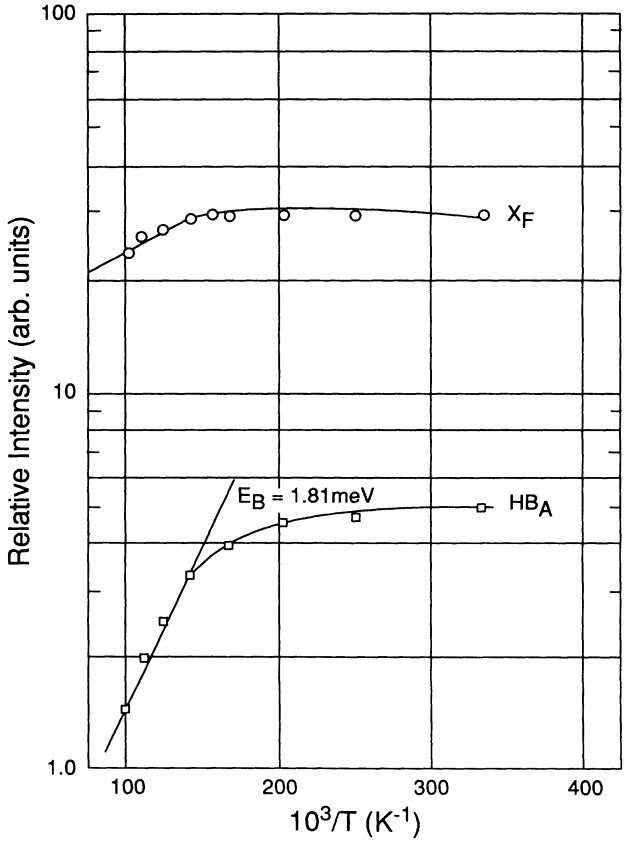

FIG. 8. The temperature dependence of the emission intensity of HB_A.

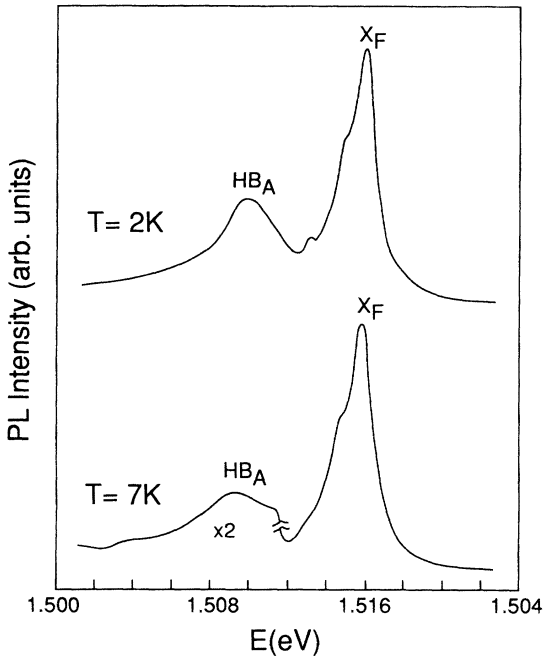

FIG. 9. The relative intensities of the direct free exciton in real space and the indirect HB_A exciton at temperatures of 2 and 7 K.

hole remains in the GaAs neutral region. The exciton then recombines as the HB_A indirect exciton in real space at the lower-emission energy.

It would be expected that the HB_A (indirect exciton) would have a smaller binding energy than the free (direct) exciton due to a greater spatial separation of the electron and hole. The binding energy of HB_A was determined from the temperature dependence of its emission intensity as shown in Fig. 8. From the slope of the temperature-dependence curve a binding energy of 1.8 meV was measured. The relative intensities of the direct free exciton in real space and the indirect HB_A exciton at temperatures of 2 and 7 K are shown in Fig. 9. It is clear that HB_A has a smaller binding energy than the direct free exciton X_F. This reflects the greater spatial separation of the electron and hole in HB_A. The temperature dependence of the HB_A transition further shows that it is not a transition between a 2D electron and a photoexcited hole.

In conclusion, we have shown that the H-band A emission results from a free indirect exciton in real space, which is directly excited by a free direct exciton in real space. However, at this point we are not sure whether the direct exciton simply distorts to become the indirect exciton or whether the direct exciton excites a completely independent indirect exciton.

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