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High angular-momentum excitons in GaAs/$Ga_{1-x}Al_xAs$ quantum wells

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We present photoluminescence excitation spectra in undoped GaAs/$Ga_{1-x}Al_xAs$ quantum wells in the presence of magnetic and electric fields perpendicular to the layers. The use of circularly polarized light allows us to separate the Zeeman components of the excitons and resolve closely lying lines in the spectra. The oscillator strength of the excited states of the heavy-hole exciton is increased by electric-field-induced coupling with the ground state of the light-hole exciton. By comparison with effective-mass-type calculations which take into account valence-band mixing and electric and magnetic-field effects, we are able to assign the peaks up to the 3d excited state.

Fine structure in the excitonic spectrum of GaAs/$Ga_{1-x}Al_xAs$ quantum wells has become resolvable in the last years, mainly due to the better quality of the samples grown by epitaxial techniques. The first excited state of the heavy-hole exciton $h_1^{(2s)}$ can be observed almost routinely in high-quality samples by photoluminescence and photoluminescence excitation[1,2] (PLE). The observation of the 3*s* excited states of the heavy- and light-hole excitons has also been reported.[3] Excited states with higher angular momenta may be observed by coupling with an *s*-type exciton.[4-6] We have recently resolved the 2*p* state of the heavy-hole exciton using the dependence of the Stark effect on the effective masses of the holes.[7] Our assignment was confirmed by calculations of exciton mixing in quantum wells.[8] These calculations also predicted that the 3*d* state of the heavy-hole exciton ($h_1^{(3d)}$) should be observable, as a consequence of its interaction with the ground state of the light-hole exciton ($l_1^{(1s)}$).

In this Rapid Communication we report the first observation of several excited states of the heavy-hole exciton with angular momentum $m \geq 1$. In the following, we shall denote these states as $h_1^{(x)}$, where x can be 2*p*, 3*p*, 3*d*, or 4*d*. The combination of three experimental tools; an electric field, a small magnetic field, and the use of circularly polarized light, was decisive for the resolution of these states. The electric field tunes the energy of the excitons, decreasing the splitting between heavy and light excitons, and allows the observation of heavy states with $m \neq 0$ through mixing of these states with the $m = 0$, $1_1^{(1s)}$ state (the electric field is not necessary for the mixing between 3*d* heavy states and $1_1^{(1s)}$, but it enhances the coupling and makes these states resolvable). The small magnetic field, 0.5 T, confines the excitons in the plane of the wells, therefore enhancing their oscillator strengths, and removes the Kramers degeneracy. The latter effect, combined with the use of σ^+ and σ^- polarized light, which selects excitons associated to holes with spin-up and spin-down components, respectively, enables the resolution of closely lying states (~0.2 meV).

The sample used in these experiments was a *p-i-n* heterostructure, consisting of five isolated GaAs quantum wells (160 Å) sandwiched between n^+ and p^+-GaAs layers. Further details of the sample were given in a previous publication.[7] The electric and magnetic fields were applied in the direction perpendicular to the layers. PLE spectra were recorded with circularly polarized light from an LD700 dye, pumped by a Kr^+-ion laser, at a temperature of 2 K. The exceptional quality of this heterostructure (typical full width at half maximum of the peaks ~0.35 meV) was a key factor for the success of the experiments.

Figure 1 shows, for σ^+ and σ^- polarizations, PLE spectra in the energy region of $l_1^{(1s)}$ and $h_1^{(x)}$ for several electric fields indicated at the right side of the spectra. At zero field, two peaks, labeled (○) and (+), are resolved in the spectra; they correspond to $l_1^{(1s)}$ and $h_1^{(2s)}$, respectively. We should mention that the electric field is not strictly zero, as may be inferred by the presence of $h_1^{(2p)}$ (●) in

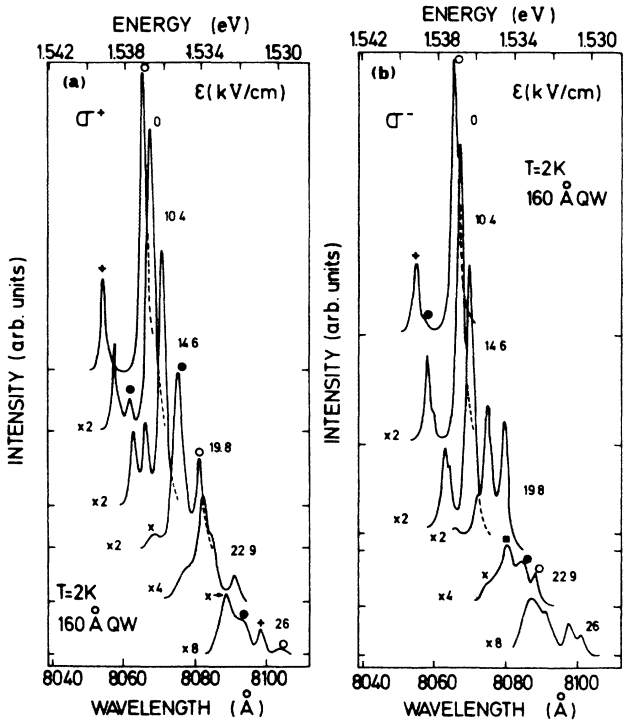

FIG. 1. Low-temperature photoluminescence excitation spectra of a 160-Å quantum well at several electric fields normal to the well. The excitation was performed with σ^+ and σ^- polarizations in (a) and (b), respectively. Scaling factors are indicated on the left-hand side of the spectra. A magnetic field of 0.5 T was applied parallel to the electric field.

σ^-, and the additional observation of the forbidden h_{12a} transition[9] (not shown in the figure). As the electric field is increased, the separation between $h_1^{(x)}$ and l_1 decreases, thereby enhancing the interaction between them. The mechanism for this interaction is different for the different states and will be explained below. The $h_1^{(x)}$ states borrow their oscillator strength from l_1, as they approach it, and become observable in the spectra. At a given field, striking differences are seen between both polarizations. For example, at 14.6 kV/cm, in σ^+, $h_1^{(2p-)}$ is clearly resolved as a peak between l_1 and $h_1^{(2s)}$, whereas, in σ^- this structure is only seen as a shoulder, $h_1^{(2p+)}$, in the low-energy side of $h_1^{(2s)}$.

Table I compiles the resolved excitonic states, and their labeling, for both circular polarizations. Using the convention of Ref. 10 (Table II), excitonic states are labeled according to the irreducible representations of the direct product of hole Bloch and exciton envelope functions, with an additional superscript indicating their ordering in energy. The main character of the excitons (heavy/light and quantum numbers of the radial functions) are tabulated in the low- and high-field limits. The symbols in parentheses refer to the resolved states shown in Figs. 1 and 2. The states are joined by a brace when a definite identification of the excitonic character was not possible. For instance, we cannot unambiguously classify the points depicted by crosses (×) in the σ^- configuration as $h_1^{(3p+)}$ or $h_1^{(4d-)}$. The ambiguity arises from the electric-field-induced broadening of the states, at fields where their oscillator strength becomes noticeable (~ 20 kV/cm), and the resulting difficulty to resolve these two closely lying states (from the calculations their splitting should be ~0.4 meV). The $h_1^{(3s)}$, not compiled in the table, lies outside the spectral range of the experiments at low fields. It enters the experimental window at high fields but could not be observed because of its small oscillator strength; however, it has been resolved in PLE experiments at ~0.7 T in the presence of a small electric field.[11]

The energy shifts and oscillator strengths of the excitonic states are plotted in Fig. 2 as a function of the electric field for both light polarizations. The points corresponding to the ground state of the heavy-hole exciton $(h_1^{(1s)},\Gamma_7^{(1)})$ were obtained directly from photoluminescence spectra, while the rest of the points correspond to the structures in the PLE spectra. The lines represent our theoretical results, using the method and parameters as in Ref. 10. In order to obtain the best agreement between theory and experiment for $l_1^{(1s)}$ and the excited states of the heavy-hole exciton, at zero field, the experimental points have been rigidly shifted by 2.7 meV towards lower energies. The reason for this discrepancy is presently not

TABLE I. Main character of the excitonic states in the low and high electric field limits for both circular polarizations. The symbols in parentheses correspond to those used in Figs. 1 and 2.

σ^+					σ^-				
Low field			High field		Low field			High field	
$\Gamma_7^{(1)}$	$h_1^{(1s)}$	(△)	$h_1^{(1s)}$	(△)	$\Gamma_7^{(1)}$	$h_1^{(1s)}$	(△)	$h_1^{(1s)}$	(△)
$\Gamma_6^{(1)}$	$l_1^{(1s)}$	(○)	$h_1^{(2p-)}$	(○)	$\Gamma_6^{(1)}$	$l_1^{(1s)}$	(○)	$h_1^{(2p+)}$	(○)
$\Gamma_7^{(2)}$	$h_1^{(2s)}$	(+)	$h_1^{(2s)}$	(+)	$\Gamma_7^{(2)}$	$h_1^{(2s)}$	(+)	$h_1^{(2s)}$	(●)
$\Gamma_6^{(2)}$	$h_1^{(2p-)}$	(●)	$h_1^{(3p-)}$	(●)	$\Gamma_6^{(2)}$	$h_1^{(2p+)}$	(●)	$h_1^{(3d-)}$	
$\Gamma_6^{(3)}$	$h_1^{(3p-)}$	(×)	$h_1^{(3d+)}$		$\Gamma_6^{(3)}$	$h_1^{(3d-)}$	(■)	$h_1^{(3p+)}$	(■)
$\Gamma_6^{(4)}$	$h_1^{(3d+)}$		$l_1^{(1s)}$	(×)	$\Gamma_6^{(4)}$	$h_1^{(3p+)}$	(×)	$h_1^{(4d-)}$	
					$\Gamma_6^{(5)}$	$h_1^{(4d-)}$		$l_1^{(1s)}$	(×)

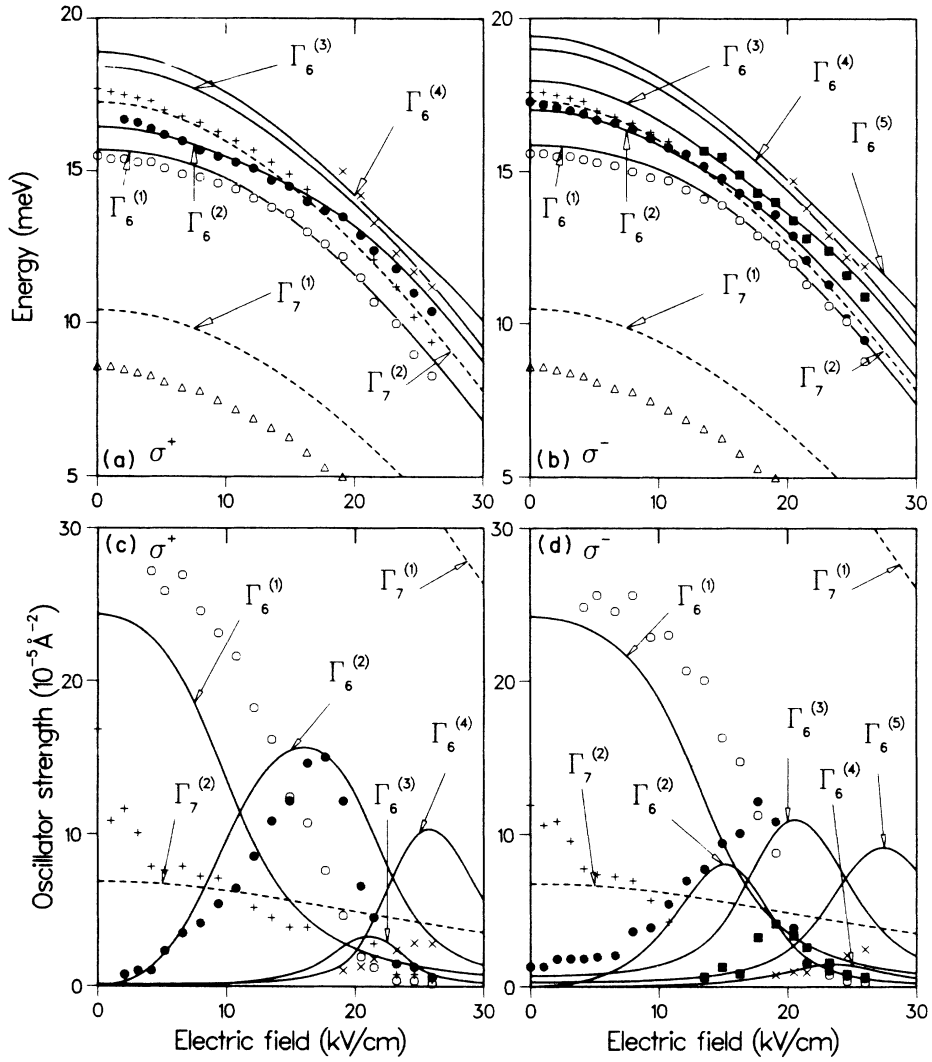

FIG. 2. Energy shifts, (a) and (b), of excitons in a 160-Å quantum well as a function of electric field normal to the well, for σ^+ and σ^- polarizations, respectively. A magnetic field of 0.5 T was applied parallel to the electric field. Oscillator strengths, (c) and (d), corresponding to (a) and (b), respectively. The lines represent the results of the calculations. See Table I for the labeling of the states.

understood. The experimental findings at low electric fields are reproduced remarkably well by the calculations: the Zeeman splitting of $h_1^{(1s)}$ and $h_1^{(2s)}$ is negligible, while that of $l_1^{(1s)}\downarrow$- $l_1^{(1s)}\uparrow$ and $h_1^{(2p+)}\downarrow$- $h_1^{(2p-)}\uparrow$ amounts to 0.15 meV, 0.5 meV in experiment (0.18 meV, 0.6 meV in theory), respectively. It is also worthwhile to emphasize the good agreement concerning the difference in magnitude of the Zeeman splitting of $m=0$ heavy and light states. The fact that this splitting is larger for light than for heavy states can only be explained if exciton mixing (nondiagonal terms in the Hamiltonian) is taken into account.[12]

As the electric field is increased, $h_1^{(2p)}$ moves closer to $l_1^{(1s)}$, these states become strongly mixed and share their oscillators strengths. The mixing is stronger in the σ^+ configuration, as can be seen from the oscillator strength and from the minimum separation between these structures: 1.2 meV in σ^+ versus 1 meV in σ^- at $\sim$18 kV/cm. In σ^-, two new structures (■ and ×) become resolvable at $\sim$13 kV/cm and $\sim$20 kV/cm, respectively. Their oscillator strengths show a similar behavior with field to that of $\Gamma_6^{(2)}$. By comparison with the theory, we assign the former structure to $h_1^{(3d-)}$, while we are not able to distinguish between the $h_1^{(3p+)}$ or $h_1^{(4d-)}$ character of the latter. In σ^+ only one additional structure (×) becomes resolvable at $\sim$18 kV/cm. Here, again, because of the closeness of $\Gamma_6^{(3)}$ and $\Gamma_6^{(4)}$, we cannot decide between its $h_1^{(3p-)}$ or $h_1^{(3d+)}$ character. In both cases, the larger oscillator strengths obtained in the calculations favor the d character, but we should mention that these strengths are quite sensitive to the choice of the band-structure parameters, especially to the value of K, which determines the effective g factor of the holes. In the high-field limit, the excited states of the heavy-hole exciton lose again their oscillator strength and will lie below the ground state of the light-hole exciton, reversing the original ordering.[7,8]

The first excited state of the heavy-hole exciton, $h_1^{(2s)}(\Gamma_7^{(2)})$, does not interact with the other states, as can be clearly seen in Figs. 2(a) and 2(b), where this state crosses the $\Gamma_6^{(2)}$ state. The calculations show that the oscillator strength of this state should decrease between 0 and 30 kV/cm only by a factor of ~ 2 [see Figs. 2(c) and 2(d)]. However, experimentally this decrease is one order of magnitude larger. One should keep in mind that, in PLE experiments, the measured magnitude is the emission of the heavy-hole exciton, and not directly the absorption of the excitonic states. The oscillator strength of $h_1^{(1s)}$ is also expected to decrease with increasing field [see $\Gamma_7^{(1)}$ in Figs. 2(c) and 2(d)]. This fact, together with the possible presence of nonradiative channels, may explain the discrepancy between calculated and experimentally obtained oscillator strengths at intermediate and high fields.

For the sake of clarity, let us discuss the mechanisms of the coupling between $h_1^{(x)}$ and $l_1^{(1s)}$ in a four-level model: two heavy states and two light states will be considered and the spin direction will be neglected in the discussion (only states observed in a given polarization may interact with each other). In this picture, the coupling between $h_1^{(2p)}$ and $l_1^{(1s)}$ takes place through $h_2^{(2p)}$, as an intermediate state. This process may be considered in two steps: the state derived from the second heavy subband $h_2^{(2p)}$ mixes with $l_1^{(1s)}$ through the lowering operator k^- [B term of Hamiltonian (5) in Ref. 10]. The electric field couples both heavy subbands, $h_2^{(2p)}$ with $h_1^{(2p)}$, therefore leading to a mixing of $h_1^{(2p)}$ with $l_1^{(1s)}$. The interaction is enhanced by the electric field, which (a) decreases the separation between $h_1^{(2p)}$ and $l_1^{(1s)}$ and (b) increases the coupling of $h_2^{(2p)}$ with $h_1^{(2p)}$. (An additional equivalent path may also be considered: the field couples $l_1^{(1s)}$ with $l_2^{(1s)}$, which is then coupled to $h_1^{(2p)}$ by k^-.) The situation is different for $h_1^{(3d)}$. The $l_1^{(1s)}$ and $h_1^{(3d)}$ states may already couple in the absence of an electric field,[8] since they belong to the same irreducible representation (Γ_{6g}) of D_{4h}. Their mixing takes place through a term containing square powers of k^- [C term of Hamiltonian (5) in Ref. 10]. In this simplified picture for the $h_1^{(3d)}$ state, the electric field mainly enhances the coupling through the tuning of the states.

In conclusion, we have resolved excited states of the heavy-hole exciton, with angular momentum higher than one, in GaAs/$Ga_{1-x}Al_xAs$ quantum wells. The observation has been possible with the combination of an electric field, to couple light and heavy states, and with the use of a magnetic field and circularly polarized light, to remove degeneracies and resolve closely lying states. The states have been identified by comparison with calculations of the effects of electric and magnetic fields on the properties of the excitons. Previous experimental uncertainties[7] about the coupling between $h_1^{(2s)}$ and $h_1^{(2p)}$ have been clarified. These results contribute to a more comprehensive understanding of the excitonic spectrum and clarify the important role of excitonic mixing in the optical properties of quasi-two-dimensional systems.

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