Term spectrum of magnetoexcitons in quasi-two-dimensional systems

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We present highly resolved photoluminescence excitation spectra of undoped $GaAs/Ga_{1-x}Al_xAs$ quantum wells in the presence of magnetic and electric fields. The high quality of our p^+-i-n^+ sample, and the excitation with circularly polarized light, allow us, for the first time, to fully resolve the fine structure in the spectra. By comparison with calculations, which takes into account exciton mixing and external fields, we identify the ground and excited states of excitons in quasi-two-dimensional systems and are able to disclose its internal structure to a hitherto unprecedented detail.

The problem of the energy-level structure of two oppositely charged particles in the presence of external fields is common to astrophysics, atomic physics, and solid-state physics. Studies of atom-field interactions have provided important keys in understanding spin-dependent interactions and angular momentum coupling schemes in atoms.[1] An early success of the optical studies of semiconductors was the demonstration of the analogy of excitons in weakly polar semiconductors with the hydrogen atom (Wannier excitons).[2] The effects of a magnetic field on the optical absorption of bulk and quasi-two-dimensional semiconductors have been extensively studied both experimentally[3-7] and theoretically[8-14] in the past. Due to the lack of samples of sufficiently high-optical quality and the theoretical machinery to calculate exciton-excited states (except in the high-field limit[9]), exciton spectroscopy in solids has not yet attained the level of sophistication routinely achieved in atomic physics. Semiconductor quantum wells (QW's), whose optical properties are strongly influenced by excitonic effects,[15] provide an ideal system to investigate two-particle bound states. An external magnetic field is an invaluable tool for these studies, since exciton oscillator strengths are enhanced and broad continuum (Fano) resonances are split into discrete lines, each corresponding to an exciton-excited state.[12] Previous magneto-optical studies in $GaAs/Ga_{1-x}Al_xAs$ QW's have been hampered by relatively large linewidths, such that the fine structure discussed later remained unnoticed. Still, useful information

regarding ground-state binding energy and band-structure parameters could be obtained.[16]

Strong effects of external fields on atomic spectra with present-day experimental facilities can be seen in highly excited states (Rydberg atoms). Indeed our experiments show many similarities with the problem of modulations in the absorption spectrum of Rydberg atoms in the presence of a magnetic field, known as "quasi-Landau resonances."[17] In the strong-field regime the states associated with the dominant lines may be assumed to be localized in the $z=0$ plane, and therefore the problem is reduced to a two-dimensional one, which then can be treated semiclassically. This approximation, however, prevents the explanation of many secondary lines present in the spectra.[1] Due to the small effective masses and strong screening of the electron-hole interaction, similar effects can be achieved in exciton spectra already in the low- and intermediate-field regime.

The fine structure of absorption spectra in a magnetic field reflects the fundamental symmetries and interactions governing excitonic states, as demonstrated previously by the observation of excited states with angular momentum higher than 1.[18] In this paper we present completely resolved exciton-term spectra in a high-quality $GaAs/Ga_{1-x}Al_xAs$ QW from low- (0.5 T) up to high-magnetic field (17 T). The spectra can be understood only by comparison with elaborate calculations, which take into account the effects of external fields, the complexity of the valence-band structure, and which realisti-

cally treat confinement effects. We present, for the first time, a direct comparison of experiments with such a theory,[14] which has been proven to work satisfactorily in the case of excitonic mixing in the presence of electric fields.[18,19] We are able to identify the lines in the spectra and to account quantitatively for the magnetic-field dependence of the energies and oscillator strengths of the excitons. Compared to previous attempts to relate theory with experiment,[11-13] we believe that the present work represents a qualitative advancement of our understanding of the optical processes in semiconductors.

Photoluminescence excitation (PLE) spectra were obtained in a polyhelix resistive magnet with fields applied in the Faraday configuration. The spectra were recorded with circularly polarized light from an LD700 dye laser, pumped by a Kr^+-ion laser, at a temperature of 2 K, with the spectrometer set at the peak of the heavy-hole luminescence, which shifts slightly with increasing magnetic field. The luminescence was normalized in real time by measuring the intensity of the exciting light, deflected through a beam splitter, with a silicon photodiode. The sample was a p^+-i-n^+ $GaAs/Ga_{0.65}Al_{0.35}As$ heterostructure with five isolated 160-Å QW's in the intrinsic region, and it has been described previously.[20]

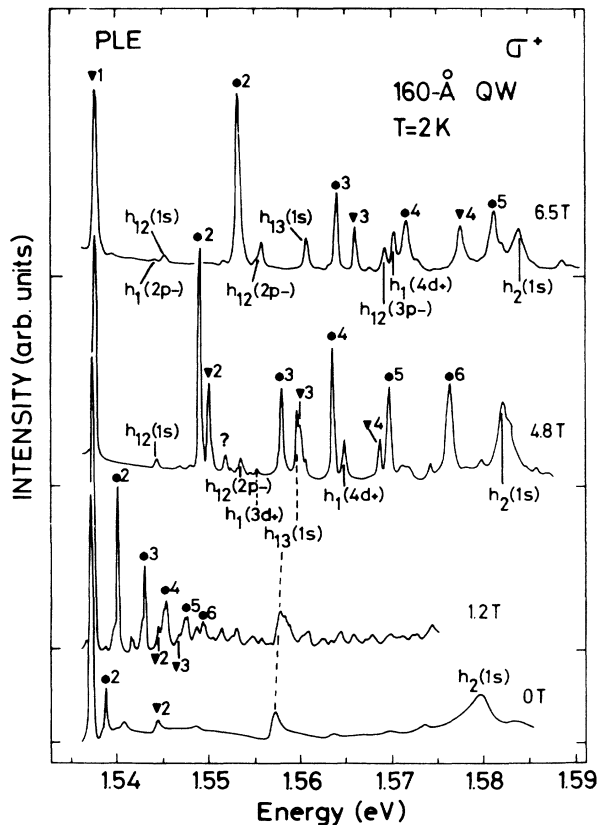Figure 1 depicts low-temperature exciton spectra, at

FIG. 1. Low-temperature photoluminescence excitation spectra of a 160-Å quantum well at several magnetic fields normal to the well. The excitation was performed with σ^+ polarization. An electric field of ~5 kV/cm is present in the n-i-p heterostructure. The ns series of h_1 and l_1 are marked by n● and n▼, respectively.

different magnetic fields, recorded with σ^+-polarized light. The zero-field pseudoabsorption consists of "allowed" and "forbidden" excitonic transitions, superimposed on a steplike background typical of two-dimensional systems. The forbidden transitions are seen because of the presence of a residual electric field of ~5kV/cm.[21] We shall use the following notation to label the lines: h (l) means heavy (light) hole; a subindex indicates the same confined subband for electrons and holes; in the case of two subindices the former corresponds to electrons and the latter to holes; a hydrogenic label, nm is used for the exciton envelope functions, where the angular momentum quantum numbers m are denoted as $(\cdots,d-,p-,s,p+,d+,\cdots)$. The spin labels may be omitted here without causing confusion.[14] The main structures in this spectrum correspond to the ground state of the light-hole exciton [$l_1(1s)$, 1.538 eV], the first excited states of the heavy-hole exciton [$h_1(2s)$, 1.539 eV] and the light hole exciton [$l_1(2s)$, 1.545 eV], the $h_{13}(1s)$ exciton [1.558 eV], and the $h_2(1s)$ exciton [1.58 eV]. The remaining peaks correspond to $h_{12}(1s)$ (1.541 eV) (h_{12a} in Ref. 22), $h_{12}(2p)$ (1.549 eV), $h_{13}(2s)$ (1.564 eV), $h_{21}(1s)$ (1.574 eV), and $h_{14}(1s)$ (1.57 eV). The heavy-hole exciton $h_1(1s)$ (not shown) was measured directly by photoluminescence. These labels are based on the calculations to be described later.

It was seen in Fig. 1 that new peaks appear in the region of the continuous absorption when a magnetic field is applied, and the density of states between the peaks vanishes. With increasing field there is a reduction of the exciton radius and a concomitant increase in the binding energy, which favors the observation of the excited states. The strongest features correspond to the ns levels of the h_1 and l_1 excitons, labeled with n● and n▼, respectively, except in some cases when they interact with other excitonic states and share with them their oscillator strength. Up to the highest field shown in the figure, the normalized intensity of $h_1(2s)$ increases monotonically with increasing field. However, this is not the case for other states, since many mutual interactions lead to a complicated behavior of their intensities. See, for example, that $h_1(4s)$ (●4) has a larger oscillator strength than $h_1(3s)$ (●3) at 4.8 T, while it is smaller again at 6.5 T. At this field the 2s state of l_1 (2▼) coincides with $h_1(2s)$ and is not seen in the spectrum.

The present effective-mass theory of magnetoexcitons in QW's has been described in Ref. 14. Briefly, the exciton envelope function is expanded into a symmetrized basis set of hydrogenic radial wave functions parallel to the interfaces and square-well subband wave functions normal to the well. The conduction band is assumed parabolic and the kinetic energy of the hole is described by the Luttinger[23] Hamiltonian. The latter contains nondiagonal terms that mix the excitons associated to the Bloch waves at the top of the valence band. Deviations from axial symmetry ("warping") are explicitly taken care of for the σ^+ polarization, where its effects should be easiest to detect. Band parameters and basis set are identical to those in Ref. 14. An electric field of 5 kV/cm is also taken into account.

Figure 2 shows the calculated and measured shifts of the excitons as a function of magnetic field for σ^+ (*a* and *b*) and σ^- (*c* and *d*) polarizations between 0.5 and 2.5 T. The energies are measured relative to the band gap of GaAs, with the experimental points rigidly shifted by 2.7 meV towards lower energies, and only a reduced range from 15 to 30 meV is shown. The area of the points is proportional to the calculated and observed intensities [normalized to the intensity of $l_1(1s)$, which increases

only weakly with field]. In the calculated pictures, the shaded areas correspond to regions where a plethora of states develop and only the main resonances $l_1(3s)$ and $h_{12}(2p\pm)$ are shown. The overall agreement between theory and experiment is very good: high-angular-momentum states such as $h_1(3p)$, $h_1(3d)$, $h_1(4f)$, and $l_1(3d)$ are identified in the experiments; the negative diamagnetic shift of $l_1(1s)$ in σ^+ and of $h_{12}(1s)$ in σ^- is
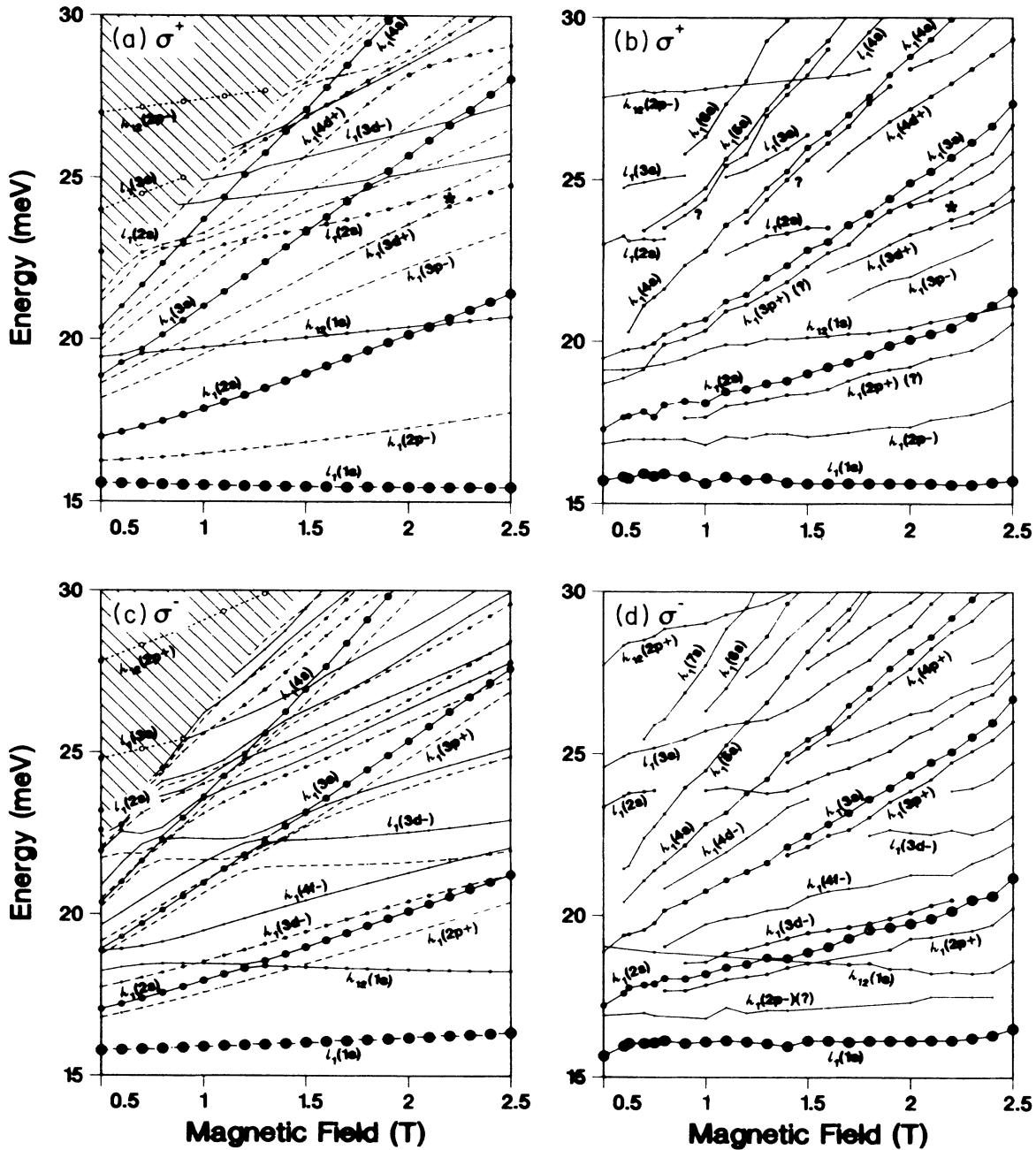

FIG. 2. Calculated [(a) and (c)] and measured [(b) and (d)] energy shifts of excitons in a 160-Å quantum well as a function of magnetic field normal to the well (for σ^+ and σ^- polarizations, respectively). The area of the points is proportional to the oscillator strengths, which are normalized to those of $l_1(1s)$. The asterisk points out the coupling between $l_1(2s)$ and $h_1(3d+)$, which is shown in Fig. 3.

also predicted by theory; even the oscillator strengths agree very well. Except at obvious anticrossings, it is often possible to label the peaks according to the predominant character of the exciton envelope function. However, this does not always hold: the dashed lines in the upper quadrant of Fig. 2(c) represent a strong mixture of $l_1(s)$, $h_1(d-)$, and $l_1(p-)$ states, which cannot unambiguously be disentangled; thus we do not assign any label to those lines in Fig. 2(d). Small discrepancies of the order of 3 meV in the energy of highly excited states as, e.g., in $h_1(4s)$, could be caused by nonparabolicity of the conduction band. More intriguing is the presence of $h_1(2p-)$ and $h_1(2p+)$ with comparable intensities in both polarizations; symmetry arguments predict that $h_1(2p-)$ and $h_1(2p+)$ should be observed only in σ^+ and σ^-, respectively. The breaking of this selection rule is presently not understood. There are also satellite lines, marked with a "?" in Figs. 2(b) and 2(d), accompanying the $h_1(ns)$ series, which are not predicted by the theory. Some of them could originate from binding of excitons to remote impurities, lattice step at the interfaces, or from spin relaxation in the cooling process of the carriers before the emission is detected, but their origin is not well established.

In Figs. 2(a) and 2(c) the solid (dashed) lines correspond to excitons with $\Gamma_7(\Gamma_6)$ symmetry in the convention of Ref. 14. States belonging to the same irreducible representation can couple and, since light-hole excitons have a smaller diamagnetic shift than heavy-hole ones and the shift increases rapidly with increasing principal quantum number n, many interactions are expected in the spectra. These predictions are borne out by our observation of clear repulsions between numerous energy levels. The determination of whether or not energy levels cross provides a sensitive test to the existence of a constant of motion in the problem: only the group $\Gamma_7(\Gamma_6)$ remains a good label and it is not possible to associate a higher symmetry to the excitons because they are of mixed character. This is, for example, the case of $l_1(2s)$ and $h_1(3d+)$ in σ^+ polarization close to 2.5 T (marked with an asterisk in the figure). This anticrossing is depicted in Fig. 3 for fields between 0 and 4 T. Both states belong to Γ_6 symmetry and can interact. As the field is increased, $h_1(3d+)$ approaches $l_1(2s)$ and gains intensity. In Fig. 3(a) the points are shown as triangles in the region of strong interaction,where they repel each other and share oscillator strength as demonstrated in Fig. 3(b). The asymmetry in the oscillator strength of $h_1(3d+)$ is due to a new interaction with $h_{12}(2p-)$ at a field of ~ 4 T. Many couplings of this type are observed in the fan diagrams, particularly in the σ^- configuration. For a given magnetic field and photon-energy range, many states are made conspicuous by borrowing of oscillator strength from neighboring excitons, which suggests that the appearance of any particular resonance may be governed by the optical excitation route employed. This would lead to stochastic processes, which are known to play an important role in atomic spectra.[24] In the energy and/or field regime (shaded in Fig. 2) where any theoretical approach similar to ours quickly becomes hopelessly complicated, an analysis of our data along these lines is expected to be more appropriate.

Finally, let us mention that at higher-magnetic fields the spectra become simpler, resembling the spectra of Landau interband transitions.[16] Excitonic effects remain quite important, however, and many interactions between excitons are still observed, though the qualitative agreement with theory somewhat deteriorates due to basis set limitations.[14] A full report of the results up to 17 T will be presented elsewhere.

In conclusion, by high-resolution experiments we were able to demonstrate that at nonzero magnetic field the absorption spectrum of quantum wells is composed of well-defined, discrete, excitonic lines, which reveal the term structure of hydrogenic pair states in semiconductors. Our observations are already obtained at low-magnetic
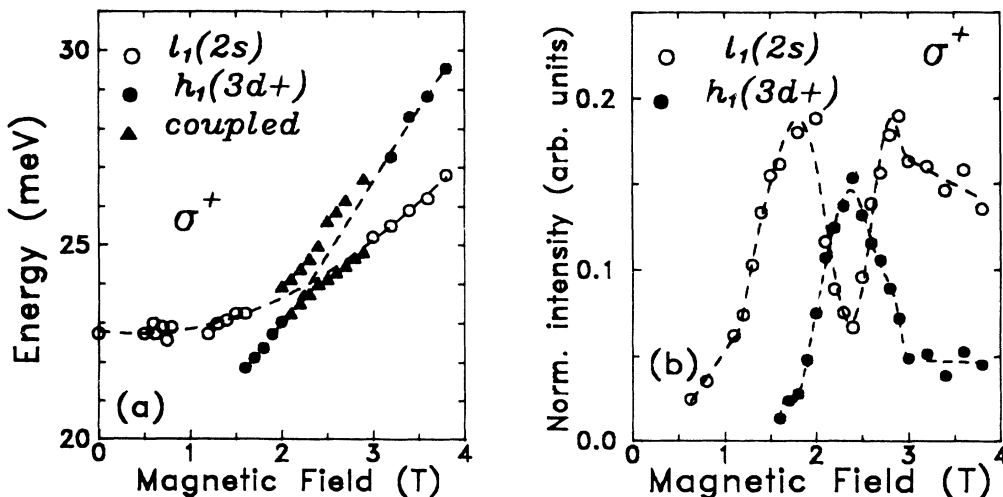

FIG. 3. (a) Energy shifts and (b) oscillator strengths of $l_1(2s)$ and $h_1(3d+)$ as a function of magnetic field in σ^+ polarization. The states are depicted by triangles in (a) in the region of strong interaction. The oscillator strengths are normalized to those of $l_1(1s)$.

fields, attainable easily in the laboratory, without the need to prepare very highly excited (Rydberg) atomic states. The comparison with theory enables the labeling of the levels and the establishment of the symmetry of the wave functions. A satisfactory agreement between theory and experiment for both energies and oscillator strengths is found. We have thus obtained strong evidence that exciton mixing in GaAs is exclusively caused by the symmetry of the Bloch functions at the fundamental band gap, i.e., that other mechanisms like, e.g., dipole-dipole[25] or exchange interactions do not play a significant role. The understanding of the intrinsic energy spectrum of excitons is indispensable for the further development of ex-

citonic physics, such as high-density excitons, exciton polaritons, bound excitons, etc., and is also relevant to many other fields of spectroscopy.

ACKNOWLEDGMENTS

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