Observation of a Magnetic-Field-Induced Transition in the Behavior of Extremely Shallow Quantum Well Excitons

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Using magnetoabsorption and electroabsorption techniques, we study excitons with binding energy comparable to the depth of GaAs/AlGaAs quantum wells. In-plane magnetic fields decrease the exciton linewidth considerably. Without magnetic fields, electroabsorption reveals 3D-like exciton behavior. This broadening persists with magnetic fields along the growth direction, but changes to a redshift for in-plane magnetic fields, reminiscent of 2D-like excitons. We discuss how in-plane magnetic fields change the nature of the exciton by inducing a two-body velocity-dependent interaction.

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A new confinement regime arises when the depth of a quantum well (QW) becomes smaller than or comparable to the exciton Coulomb binding energy [1,2]. A number of systems, including small band offset II-VI [3] and III-V [1,2] semiconductor wells and wires, lie in this relatively unexplored intermediate regime between 3D- and 2D-like exciton behavior [1,4]. In the 3D limit, the electron (e) and hole (h) motions are strongly correlated in all directions, but the two-body exciton problem can still be described in terms of *uncorrelated* relative (RM) and center of mass (CM) motions. As the well depth increases, the breaking of translational invariance invalidates this picture and eventually leads to 2D-like exciton behavior in deep and narrow wells [5]. In this case, the e and h move along the growth direction *approximately independent* of each other and, to first approximation, may be thought of as *separately confined* by the bare well potential [5]. In this Letter, we study excitons in $GaAs/Al_xGaAs_{1-x}$ multiple quantum wells (MQW's) with Al concentration $x < 2\%$. We use electroabsorption and magnetoabsorption to identify the physical mechanism that drives the transition from 3D-like towards 2D-like excitons. In-plane B fields change the nature of the exciton motion along the growth direction from that of a *strongly correlated* e-h pair (3D limit) to that of a *more weakly correlated* e-h pair. This is not the case when B points along the growth direction. Physical mechanisms that can drive such transitions are of fundamental interest [6]. Interest in shallow wells has also been spurred by their proposed use to increase switching speeds in absorption modulator devices [7]. Note that the 2D-like exciton picture, developed for deep QW's [5], breaks down in our systems [8]. Although the wells can sustain weakly confined e and h subbands, *the 3D Coulomb interaction couples them strongly to the QW excited states.*

The nature of excitons in MQW systems can be optically studied with electroabsorption. This technique probes the *changes* in the exciton absorption induced by a static electric field E_z applied along the growth direction. E_z ionizes the relative motion bound state of a 3D-like exciton. This broadens and blueshifts the exciton absorption, which is referred to as the Franz-Keldysh effect (FKE) [9]. For 2D-like excitons, the e and h are *independently confined* by the QW potential, which inhibits the ionization. This is manifested experimentally by a *redshift* in the exciton absorption peak [10]. This is the quantum-confined Stark effect (QCSE). Electroabsorption can therefore be used to study the transition from 3D- towards 2D-like excitons.

We studied $GaAs/Al_xGa_{1-x}As$ MQW's, with Al concentration varying between $x = 2\%$ and 0%. 50 periods of 100 Å GaAs wells and 100 Å Al_xGaAs_{1-x} barriers were grown by molecular beam epitaxy [2]. To apply E_z, these MQW's were placed in the intrinsic region of a p-i-n diode. The samples were patterned into 200 μm diameter mesas and contacted by wire bonding and In alloy. The built-in voltage of these p-i-n diodes was found to be $\sim$+1.5 V, determined by the applied voltage necessary to achieve flatband condition. The substrate was chemically etched off and the back surface antireflection coated for absorption measurements. Magnetic fields, both perpendicular and parallel to the QW plane, were applied using a 7 T split-coil superconducting magnet. Absorption measurements were performed using a broadband bulb filament and grating spectrometer.

Previous photocurrent measurements showed that the QCSE yields to the FKE as x decreases below $\sim$0.8% [2]. Using $\Delta E_g \sim 1237x$ (meV) for the total band offset, and assuming a conduction to valence band offset ratio of 60/40, $x = 0.8\%$ corresponds to well depths of $V_e =$ 5.94 meV (electron) and $V_h = 3.96$ meV (hole). These are *comparable* to the bulk GaAs exciton binding energy $\sim$4 meV. Within the 2D-like exciton picture, the confinement along the growth direction is approximately described by the Kronig-Penney model. This predicts a *very large* superlattice miniband width for $x = 0.8\%$ ($\sim$15 meV for the electrons), which significantly exceeds

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the above barrier heights [2]. Therefore, the observation of QCSE (implying exciton confinement by a single QW) down to $x = 0.8\%$ is quite surprising within this model. The 2D-like exciton picture, used in deep wells [5], clearly fails to describe the effects of shallow wells on the exciton. Neither the electron *nor the heavier hole* is strongly confined by the well potential alone [8]. Therefore, we cannot attribute the 2D-like exciton behavior to the mechanism studied by Wu and Nurmikko [3].

We chose a sample with $x = 0.25\%$, where at $B = 0$ the electroabsorption measurements yield FKE exciton behavior. Figure 1(a) displays a series of absorption spectra, at flatband bias (i.e., $E_z = 0$), as a function of magnetic field $B_\perp$ *perpendicular* to the QW plane. On the high energy side, a series of absorption peaks shift approximately linearly with $B_\perp$ [see inset fan plot in Fig. 1(a)] and are therefore attributed to transitions between Landau levels (LL's). The absorption-edge peak, on the other hand, exhibits a quadratic diamagnetic shift, characteristic of strongly bound excitons. With an in-plane magnetic field $B_\parallel$, however, the exciton behavior is *quite different* from the $B_\perp$ case, as Fig. 1(b) demonstrates. The exciton linewidth *narrows considerably* with increasing $B_\parallel$ and by $B_\parallel = 4$ T becomes comparable to the intrinsic linewidth in a test bulk sample with $x = 0\%$. As shown in the inset, this peak appears to display an initial small redshift, and an overall quadratic blueshift with $B_\parallel$. By $B_\parallel = 4$ T, a second peak has emerged near the band edge. This shifts quasilinearly for $B_\parallel > 4$ T (see inset), which indicates the formation of the lowest LL. Unlike with $B_\perp$, we did not observe any other significant LL-like peaks. The formation of higher LL orbits must therefore be inhibited along the growth direction, due to the disorder and the Al-induced confinement. To ensure that the above differences between $B_\perp$ and $B_\parallel$ are not due to extrinsic effects, we looked at the diamagnetic shifts in a $x = 0\%$ (bulk GaAs) test sample, grown in an identical configuration. The absorption did not depend on the B-field orientation, as expected in the absence of any preferred direction.

We now consider the electroabsorption data from the $x = 0.25\%$ sample, in the presence of magnetic fields. For $B = 0$, Fig. 2(a) demonstrates FKE behavior in this sample. This persists when we apply perpendicular magnetic fields. For in-plane magnetic fields, however, the

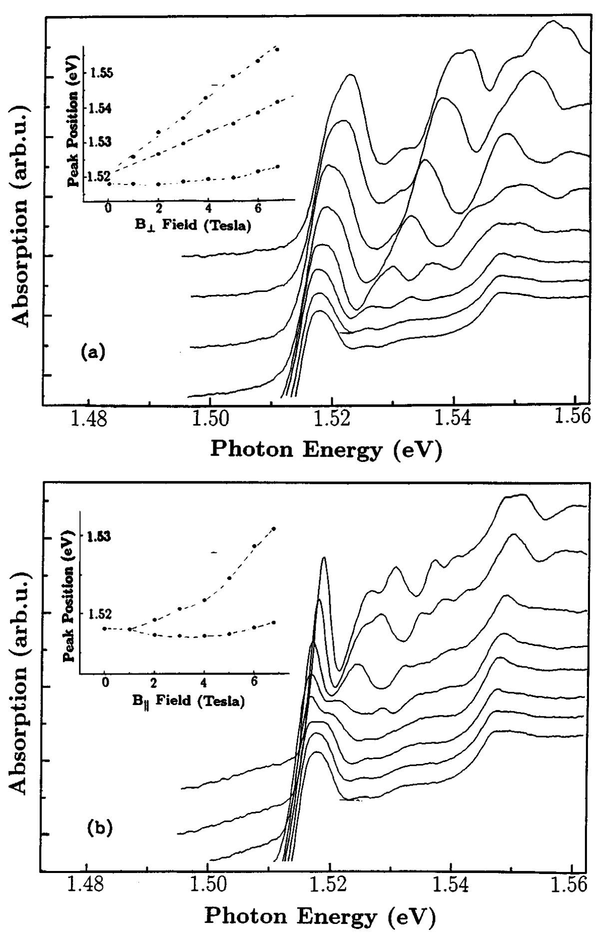

FIG. 1. Absorption spectra for $x = 0.25\%$ shallow well, as a function of $B_\perp$ (a) and $B_\parallel$ (b) (0–7 T, steps of 1 T). Insets: Fan plots of transition energies. $T = 10$ K.

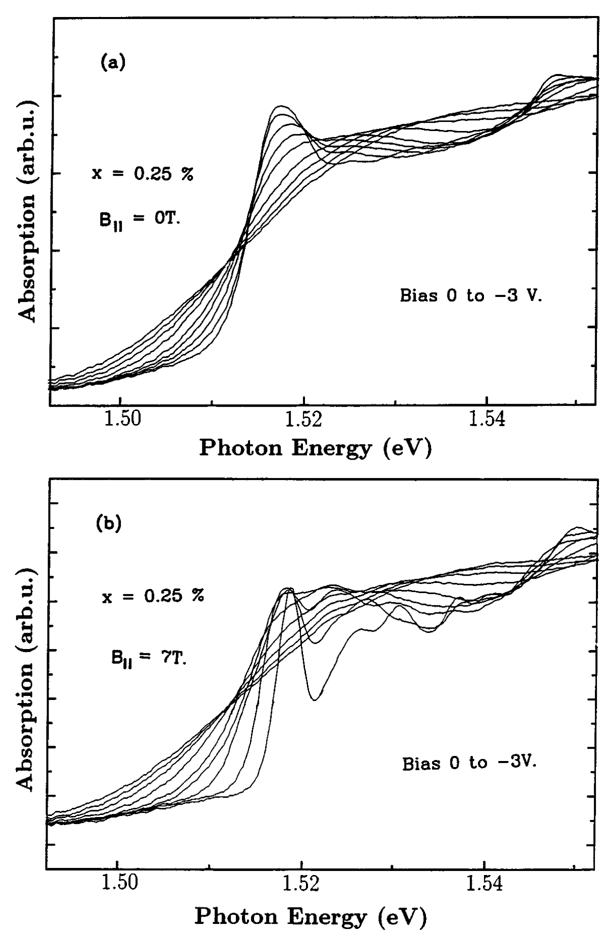

FIG. 2. Electroabsorption spectra for $x = 0.25\%$ sample, for $B_\parallel = 0$ (a) and $B_\parallel = 7$ T (b). Reverse bias ranges from 0 to -3 V. $T = 10$ K.

electroabsorption behavior is *quite different*. Figure 2(b) displays the electroabsorption data for the $x = 0.25\%$ sample, at $B_{\|} = 7$ T. A quadratic redshift with applied E field is clearly observed before the onset of exciton ionization. The latter is now *inhibited by the QW in an observable way,* unlike for $B_{\perp}$ or $B = 0$. This indicates a transition from FKE to QCSE electroabsorption behavior, unlike for $B_{\perp}$. In addition, the LL-like peak, at $\sim$1530 meV, shifts *quasilinearly* with E_z, reminiscent of coupled well or Stark ladder behavior [11].

To understand the nature of extremely shallow QW excitons, we solved the following Hamiltonian:

$$H = \frac{[\vec{p}_e + (e/c)\vec{A}(\vec{r}_e)]^2}{2m_e} + \frac{[\vec{p}_h - (e/c)\vec{A}(\vec{r}_h)]^2}{2m_h} - \frac{e^2}{\epsilon|\vec{r}|} + V_e(z_e) + V_h(z_h). \quad (1)$$

$\vec{r} = \vec{r}_e - \vec{r}_h$ denotes the e-h relative coordinates and ϵ the dielectric constant. In the Landau gauge, the vector potential is $\vec{A}(r) = \frac{1}{2}(\vec{B} \times \vec{r})$. Based on [1], we use the bulk GaAs band-structure description for excitons in extremely shallow wells. The strong mixing of the heavy and light hole valence bands is thus approximated by a single quadratic valence band with $m_h = 0.15m_0$ (m_0 is the free electron mass) and $m_e = 0.067m_0$. By increasing the Al concentration from $x = 0\%$, we break translational invariance along the z direction. We model this here with square-well potentials, $V_e(z_e)$ and $V_h(z_h)$. A standard canonical transformation [12] produces

$$H = \frac{P_Z^2}{2M} + H_{\rm RM} + H_{\rm int}\,. \quad (2)$$

For in-plane magnetic field along the y direction

$$H_{\rm RM} = \frac{p_r^2}{2\mu} + \frac{eB_{\|}}{2\mu c}\left(\frac{m_h - m_e}{m_e + m_h}\right)L_y + V(r) + \frac{e^2B_{\|}^2}{8\mu c^2}(x^2 + z^2), \quad (3)$$

L_y is the angular momentum component, and

$$H_{\rm int} = V_e\left(Z + \frac{m_h}{M}z\right) + V_h\left(Z - \frac{m_e}{M}z\right) - \frac{eB_{\|}}{cM}xP_Z\,. \quad (4)$$

In the above, capital letters describe the operators associated with the CM motion along the z direction, while lower case letters denote the operators associated with the RM. In the limit of extremely shallow wells, V_e and V_h should be thought of as inducing an effective two-body interaction between the RM and CM motions. In addition to the one-body harmonic potential in Eq. (3), $B_{\|}$ induces a *two-body velocity dependent* interaction between the CM and RM motions [last term in Eq. (4)]. This is *absent* for $B_{\perp}$, where $P_{\|} = 0$ for allowed optical transitions. In this case,

$$H_{\rm RM} = \frac{p_r^2}{2\mu} + V(r) + \frac{e^2B_{\perp}^2}{8\mu c^2}(x^2 + y^2). \quad (5)$$

We solved the Hamiltonian variationally by expanding in a real-space basis set. For $B_{\|}$, the *basis elements* were

$$(1, z_e z_h, ixz_e, ixz_h)e^{-a_j z_e^2}e^{-b_k z_h^2}e^{-c_l(x^2+z^2)}e^{-d_n y^2}. \quad (6)$$

For $B_{\perp}$, we interchanged z and y in Eq. (6) to take advantage of the symmetry. We reproduced both the quantitative results of [12] for bulk magnetoexcitons and 2D-like exciton behavior in deep wells. Note that, unlike in previous studies [3,5], *the coordinate dependence of our wave function is quite unbiased:* it is *not* separable in the in-plane (x, y) and growth (z) directions, nor in any other way, and *does not* rely on subband (or LL) expansions, which converge *very slowly* in extremely shallow wells.

Figure 3 summarizes the essence of our results (see also [8]). We focus on the nature of the exciton motion along the growth direction. $\Delta z_h = \sqrt{\langle z_h^2 \rangle}$ measures the confinement of the h when part of the exciton. For $B = 0$, we find that the exciton wave function in extremely shallow wells can be approximated by the product of the 3D RM bound state and a *confined* CM wave function [8]. This corresponds to *uncorrelated* RM and CM motions, as in the strictly 3D limit. Although the exciton can be thought of as 3D-like, its CM motion is confined. Extremely shallow wells therefore confine the exciton as a whole, rather than the e and h individually as deep wells do. FKE behavior is then expected in electroabsorption

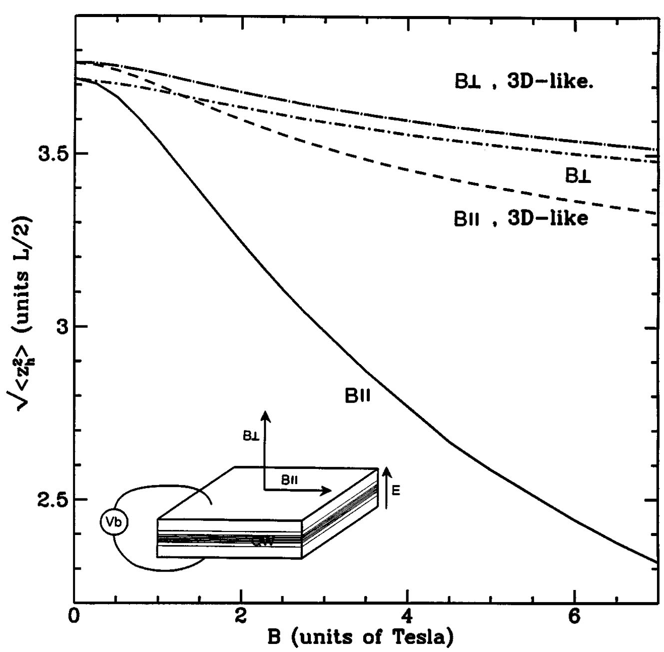

FIG. 3. Theoretical calculation of change in hole confinement with $B_{\|}$ and $B_{\perp}$. We compare to results for 3D-like excitons. $V_e = 1.2$ meV, $V_h = 0.8$ meV. Well width $L = 100$ Å.

because the wells cannot strongly inhibit the ionization of the RM bound state. The question then arises: how do magnetic fields affect such a 3D-like exciton? In the case of $B_\perp$, the harmonic magnetic potential in Eq. (5) increases the binding of the RM bound state. We should therefore expect $B_\perp$ to *enhance* the validity of the 3D exciton picture in extremely shallow wells: it would be more difficult for the wells to deform the RM bound state as $B_\perp$ increases. Figure 3 demonstrates that this is indeed the case for $B_\perp$. By comparing the h confinement to that obtained with a 3D-like variational wave function, we find that the agreement becomes better as $B_\perp$ increases. Similar behavior was obtained for all other calculated quantities [8]. With $B_\parallel$, the 3D-like picture remains valid *only if we neglect the* xP_Z *interaction* in Eq. (4). The main B effect would then be the harmonic RM potential in Eq. (3). The hole confinement thus obtained follows the 3D-like exciton curve in Fig. 3 (for $B_\parallel$) [8]. Even though the harmonic RM potential does contribute to the exciton confinement, its effect is much smaller than that of the xP_Z interaction. Figure 3 indicates that the latter *invalidates* the 3D-like exciton picture. Indeed, it *significantly reduces the e-h correlation along the z direction* [8], while *dramatically enhancing* the exciton confinement. This allows the wells to inhibit the RM ionization, leading to QCSE electroabsorption behavior.

The above experimental and theoretical results shed light into the physics behind the transition from strongly to weakly correlated e-h pair motion along the growth direction in QW systems. The harmonic magnetic potential tends to make the 3D-like exciton description more robust in extremely shallow QW's, by decreasing the size of the RM bound state. This is consistent with our $B_\perp$ results. The transition with $B_\parallel$ towards a 2D- rather than a 3D-like exciton is due to the dynamical coupling of the CM and RM motions along the growth direction, induced by the xP_Z two-body interaction in Eq. (4). The strength of such processes, describing a continual transfer of energy back and forth between the CM and RM degrees of freedom, is directly controlled experimentally by changing $B_\parallel$. This is more illuminating than simply increasing the well depth and manifests itself in electroabsorption by enhancing the QCSE. In addition, the RM harmonic potential $\propto z^2$ also contributes to the magnitude of the observed exciton redshift.

The broad linewidth of our 3D-like exciton is not surprising, in view of the large disorder fluctuations in the barrier for small Al concentrations. Figure 1 clearly demonstrates that this linewidth narrows considerably *only with in-plane magnetic field*. This is consistent with the result of our calculation that the xP_Z interaction, absent for $B_\perp$, *strongly enhances* the exciton confinement inside the impurity-free GaAs QW.

In conclusion, we studied the physical mechanism that drives the transition from 3D- towards 2D-like exciton behavior in semiconductor QW's. Extremely shallow wells confine the exciton as a whole without completely invalidating the 3D-like picture. The latter is destroyed by the dynamical coupling of the CM and RM motions, controlled by an in-plane magnetic field.

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