

## Two-Dimensional $D^-$ Centers

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Far-infrared magnetotransmission and magnetophotoconductivity measurements on selectively doped GaAs-GaAlAs multiple-quantum-well structures reveal photoionization transitions from two-dimensional  $D^-$  centers to successive Landau levels. Selective doping of the quantum-well structure favors the formation of  $D^-$  centers in the well. The two-dimensional nature of this center results in a dramatic enhancement of its binding energy with respect to the three-dimensional case.

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Two-dimensional (2D) shallow donors in semiconducting quantum wells have been the subject of considerable study recently.<sup>1-9</sup> Attention has been mainly focused on *neutral shallow donors*  $D^0$  in intentionally doped GaAs-GaAlAs multiple-quantum-well (MQW) structures.  $D^-$  centers, i.e., *neutral shallow donors that bind an additional electron*, can also be expected to readily form in this type of structure due to electron transfer from the GaAlAs barrier to neutral donors located in the GaAs well.  $D^-$  centers have already been observed in three dimensions (3D) in elemental semiconductors<sup>10,11</sup> and III-V compounds.<sup>12-14</sup> In bulk material,  $D^-$  states are populated by optimizing the experimental parameters such as band-gap radiation and the electric-field bias applied to the sample at low temperatures. In addition, very pure, low-compensation material is required, otherwise donor-acceptor or  $D^0 \rightarrow D^+$  recombination inhibit the buildup of electrons on  $D^0$  sites.<sup>13,14</sup> Thus, a "dynamic" equilibrium of photoexcited electrons trapped to  $D^0$  sites is achieved for bulk material. In comparison, recombination of  $D^-$  states in a quantum well via  $D^+$  centers in the barrier is effectively blocked by the confining potential. Thus, a "static" equilibrium of  $D^-$  states can be realized in an optimized structure whereby a neutral donor in the well would trap an additional electron, a physical situation in sharp contrast to bulk material.

Negatively charged hydrogen ions (the  $D^-$  state is the solid-state analog of the  $H^-$  ion) are of interest in many branches of physics, e.g., astrophysics.<sup>15,16</sup>  $D^-$  states confined to a quantum well opens many new interesting possibilities to study correlation effects in low-dimensional systems.

In this Letter, evidence is presented for the identification of two-dimensional (2D)  $D^-$  centers in selectively doped GaAs-GaAlAs MQW structures. These experiments represent the first observation of  $D^-$  states in confined geometries. They reveal a dramatic enhance-

ment of the binding energy due to the reduced dimension. In addition, some previously unidentified features in the transmission spectra observed by several groups, studying the same system, are explained in terms of 2D  $D^-$  state transitions.

Far-infrared transmission and photoconductivity measurements are performed on selectively silicon-doped GaAs-GaAlAs MQW structures using an optically pumped laser and a magnetic field up to 21 T. Two 100-Å-well/100-Å-barrier samples with 150 periods are studied (Al content is 25%). Samples 1 and 2 are planar doped at the middle of each well and at 10 Å before the interface, respectively, with  $10^{10}\text{-cm}^{-2}$  silicon impurities per well. The magnetic field is applied perpendicular to the interface. These samples have been studied previously by far-infrared<sup>6,7</sup> and luminescence<sup>9</sup> techniques and are therefore well characterized. There are several important points to note concerning the present set of experiments:

(i) Despite planar doping in the quantum well, there is partial segregation of the donors during growth, such that some of them are found around the center of the barrier. This is more important in sample 2 which has been doped towards the end of each quantum well.<sup>6,7,9</sup>

(ii) Because of compensation and self-compensation<sup>9</sup> (silicon is amphoteric in GaAs), some donors are ionized.

(iii) All the donors in the GaAs wells remain neutral<sup>6</sup> or they bind a second electron (see below).

(iv) The positively ionized donors are all located in the GaAlAs barriers where the binding energy is lowest.<sup>6</sup> However, there still remain neutral donors in the barriers, i.e., electrons in the well that are weakly bound to their parent ion in the barrier.<sup>4</sup> Thus, the samples have a "built-in" surplus of electrons on  $D^0$  sites in the wells. This is an ideal situation for the formation of  $D^-$  centers in the quantum well.

A magnetotransmission (MT) spectrum at  $E=17.58$

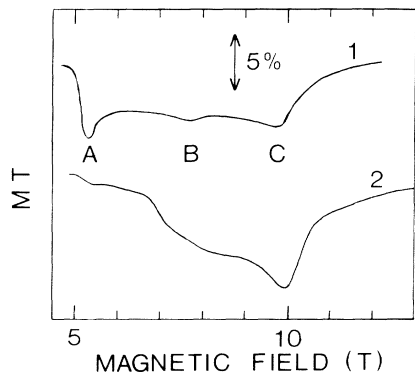


FIG. 1. Transmission spectra (arbitrary units) of samples 1 and 2 at the laser energy  $E=17.58$  meV. A 5% change in transmission is shown.  $T=4.2$  K.

meV is displayed in Fig. 1 for each of the samples. Three prominent absorption peaks can be seen. These three transitions are well documented in the literature by several independent groups<sup>4-7</sup> and are characteristic features of the transmission spectra of selectively doped GaAs-GaAlAs MQW's. All three peaks are confinement related.<sup>6,7</sup> Peaks *A* and *C* are the  $1s \rightarrow 2p^+$  transitions for donors located in the middle of the well and at the middle of the barrier, respectively. These peaks reflect the high density of states at the high-symmetry points of the MQW structure.<sup>1,2</sup> Impurities located at other places do not contribute much to the absorption signal unless they have been incorporated in a sharp doping profile. For example, the dopant spike located near the interface in sample 2 contributes to a poorly resolved feature on the low-field side of peak *B*. Careful transmission measurements at a lower field than in Fig. 1 fail to reveal additional absorption lines. There has been considerable controversy about the interpretation of the unexpected peak *B*.<sup>4-7</sup> This peak is not observed in very selectively barrier-doped samples which reveal only peak *C* while samples doped with very sharp profiles in the well reveal only peak *A*.<sup>17</sup> Peak *B* appears once donors are simultaneously present in the well and in the barrier<sup>4,6,17</sup> and, therefore, appears to be linked with the existence of an electron surplus as compared to neutral donors in the well. No conclusive identification has been assigned for this transition up to now. Below, we show that  $D^-$  centers in the quantum well are in fact responsible for this peak.

Figure 2 displays a magnetophotoconductivity (MPC) spectrum for sample 2 at the same laser energy as before. This spectrum clearly exhibits the main advantage of the photoconductivity technique in revealing weak transitions not observable in transmission. A series of oscillations (including peak *B*) quasiperiodic with the reciprocal magnetic field is observed. The line shape and periodicity of the oscillations are reminiscent of the "sawtooth" photoionization transitions from a  $D^-$  state

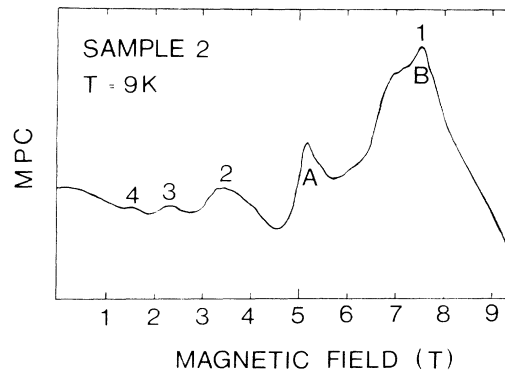


FIG. 2. Photoconductivity spectra (arbitrary units) of sample 2 at  $E=17.58$  meV. The peaks *A* and *B* (together with its low-field shoulder due to near-interface donors) are clearly seen. The photoionization transitions  $D^- \rightarrow N$  are labeled according to their final Landau level.

to Landau levels that were observed in the photoconductivity response of bulk GaAs.<sup>12-14</sup> The peak positions are displaced to a lower field in accordance with an increased binding energy due to quantum confinement. It is thus proposed that peak *B* is due to a photoionization transition from a  $D^-$  state (located in the well) to the  $N=1$  Landau level. The harmonic structure at a lower field is then due to the corresponding  $D^- \rightarrow N > 1$  photoionization transitions.<sup>18</sup> The series of oscillations of Fig. 2 is also seen in sample 1 but it is considerably weaker in accordance with a reduced surplus of electrons in the well due to smaller donor segregation in the barrier. The above assignment is substantiated by photoconductivity measurements performed at energies  $\hbar\omega_c$  ( $\hbar\omega_c$  is the cyclotron energy) less than that of peak *B* (Refs. 13 and 14) to investigate the fundamental  $D^- \rightarrow N=0$  photoionization transition. An example of the results is shown for sample 1 in Fig. 3 where a broad, symmetrical peak (not seen in transmission measure-

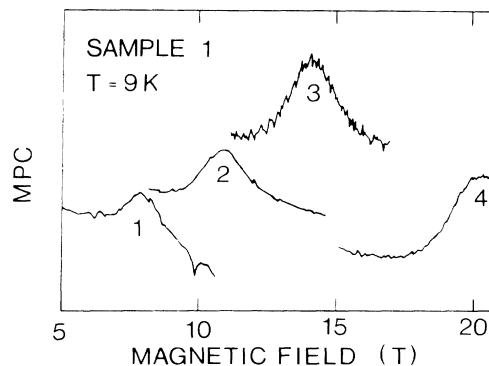


FIG. 3. Photoconductivity spectra (arbitrary units) of sample 1 showing the  $D^- \rightarrow N=0$  photoionization transition. The laser energies are 1, 5.78 meV; 2, 6.12 meV; 3, 6.72 meV; 4, 7.61 meV.

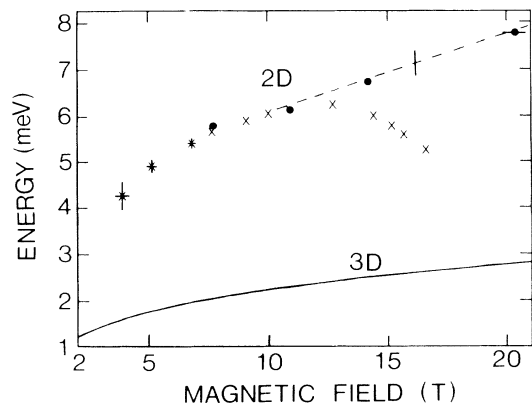


FIG. 4. Binding energy of the 2D  $D^-$  center as a function of field. The crosses are obtained by subtracting the cyclotron energy from the energy of peak  $B$ . The dashed line is obtained the same way but after correction for polaron effects as discussed in the text. The solid circles correspond to the  $D^- \rightarrow N=0$  peaks of Fig. 3. Typical error bars are indicated. For comparison, the theoretical binding energy of a 3D  $D^-$  center is shown (Ref. 21). In 3D, theory and experiment are in reasonable agreement (Ref. 14).

ments) is observed for different energies below 7.6 meV. In sample 2, this peak is more intense again because of a larger surplus of electrons in the well, but it is broader. It is worth emphasizing that in contrast with the bulk case,<sup>13,14</sup> all the spectral features due to  $D^-$  states are observed *in the absence of visible light illumination* (dark conditions).

In Ref. 7, peak  $B$  was followed over a wide range of energy up to the optical-phonon energy specifically to study resonant polaron coupling. A linear increase in energy as a function of the magnetic field was observed up to approximately 25 meV. At higher energies there is a sublinear increase with field due to polaron effects which can be subtracted by extrapolating the low-field linear results to high fields.<sup>7</sup> The energy of the  $D^- \rightarrow N=1$  transition obtained this way minus  $\hbar\omega_c$  allows for an accurate determination of the binding energy of the 2D  $D^-$  state. The result of such a procedure is shown in Fig. 4 together with the data corresponding to the fundamental  $D^- \rightarrow N=0$  transition of Fig. 3. Excellent agreement is found between the two sets of data over a wide range of magnetic field above 5 T.

To our knowledge, there is, to date, no calculation of 2D  $D^-$  states which could be used for a quantitative comparison with our data. However, there are two features of Fig. 4 which are worth commenting on: a very strong enhancement of the binding energy and a steeper field dependence with respect to the 3D case. At zero field, only one bound spin-singlet state exists for the  $D^-$  center in 3D,<sup>15</sup> with an energy of  $0.055R$  (where  $R$  is the effective Rydberg; for GaAs,  $R=5.8$  meV). This ground state is rather well described by a Chandrasekhar trial wave function which is the symmetrical product of

two  $1s$  hydrogen functions multiplied by a correlation factor. This correlation factor is crucial since it accounts for about half of the binding energy. At  $B=0$ , the "inner" orbital is almost unaffected by the weakly bound "outer" orbital which extends over approximately 400 Å. When a magnetic field is applied, the  $D^-$  orbital is squeezed to approximately the cyclotron radius in the plane perpendicular to the magnetic field but remains largely extended parallel to the field as a consequence of the electron correlation.<sup>19</sup> This produces a dramatic deepening of the ground state (typically an order of magnitude for 20 T). In addition, many more states can bind, including spin triplets.<sup>19-21</sup> By analogy to confined  $D^0$  states,<sup>1,2</sup> the strong confinement of the  $D^-$  orbital in the 100-Å quantum well will certainly result in an enhancement of the binding energy. By comparing the field dependence in 2D and 3D in Fig. 4, a zero-field binding energy of approximately 2 meV can be anticipated. This is 5 times more than in 3D. In addition, correlation effects are very important and increase with field strength. Thus, enhanced correlation effects are probably responsible for the steeper field dependence in 2D vs 3D (0.16 meV/T at high field as compared to 0.06 meV/T).

To summarize, the existence of "built-in" confined  $D^-$  centers in intentionally doped GaAs-GaAlAs MQW's is reported. These centers give rise to previously unexplained spectral features in the energy-field range of the  $1s \rightarrow 2p^+$  transitions for neutral donors. The existence of a built-in population of  $D^-$  centers opens a wide variety of physical phenomena related to negatively charged donors in a semiconductor to be studied. Indeed, the samples can be "engineered" to enhance the  $D^-$  state population. On the basis of the present experimental findings, quantitative calculations are required to elucidate the peculiar behavior of a 2D  $D^-$  center in a high magnetic field, in particular, the key role played by correlation. Also of interest is the theoretical search for possible new bound states due to the confinement and for the formation of a "spatial  $D^-$  band" by analogy to the  $D^0$  band.<sup>1</sup>

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