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Magnetic-field-induced unbinding of the off-well-center D^- singlet state in $GaAs/Al_{0.3}Ga_{0.7}As$ multiple quantum wells

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Results of far-infrared Fourier-transform magnetotransmission from two $GaAs/Al_{0.3}Ga_{0.7}As$ multiple quantum well samples δ-doped with Si-donor on- and off-well centers are compared. In contrast to well-center D^- ions, the off-well-center D^- singlet binding energy decreases with increasing magnetic field between 5.5 and 15 T, as reflected by the decreasing strength of the D^- singlet transition and a consequent increase of the neutral donor $1s$-$2p^+$ transition. This observation represents a verification of the predicted magnetic-field-induced unbinding (magnetic evaporation) of shallow impurity states for the off-well-center D^--ion system. [S0163-1829(97)50428-0]

Recently, there have been several interesting theoretical considerations of a two-electron problem in magnetic field. In a model proposed for the quantum dot, exact numerical solution of the problem of two interacting electrons in the x-y plane bound in a harmonic-oscillator potential has been reported.[1,2] These authors predicted that the two-electron ground state can oscillate between the spin-singlet and spin-triplet states as a function of the magnetic-field strength; and that for a fixed quantum dot size but with increasing magnetic field, the spin-triplet state will finally become the ground state. These unusual magnetic-field effects are a result of electron-electron interactions (correlations). However, probing of such effects in nearly parabolic quantum dots directly via far-infrared (FIR) absorption is forbidden due to a generalization of Kohn's theorem.[3–5] FIR radiation couples only to the center-of-mass motion for a parabolic confinement potential, and hence, is insensitive to electron-electron interactions. On the other hand, similar in spirit to this situation, two interacting electrons in the x-y plane bound to a fixed positive ion whose position relative to the x-y plane is variable offer a rather simple system to study electron-electron interactions directly.[6,7] For a configuration in which the positive ion is located at a distance from the x-y plane, a magnetic-field-induced progression of the bound two-electron ground state from a singlet to a triplet configuration has been described, and an unusual magnetic ''evaporation,'' i.e., magnetic-field-induced unbinding of such a D^- center, has been predicted.[7] One way to realize a similar situation is to place Si donors away from the centers of $GaAs/Al_xGa_{1-x}As$ quantum wells in which the centers of wide barriers are δ-doped with Si donors, so that donors in the wells can trap the second electron provided by barrier donors to form ''off-well-center'' D^- ions. Numerical calculations for such realizable quantum-well systems have also revealed similar magnetic-field effects.[8,9]

When donors in the wells are located more than a quarter of the well width away from the well center, the off-well-center D^- singlet binding energy initially increases, then decreases with increasing magnetic field, eventually becoming dissociated.[8] The off-well-center D^- triplet ground state exhibits similar qualitative behavior, but is predicted to dissociate at a much larger field. Therefore, a crossover between the ground singlet and triplet states occurs at some magnetic-field strength, above which the triplet state will be the ground state of the system. Such unusual magnetic-field effects are a direct result of magnetic-field tuning of the competition between the electron-electron repulsion and the attractive interactions of the electrons with the positively charged impurity center. For an off-well-center D^- ion, since the peak of the electron probability density along the growth direction is displaced from the position of the positive charge, with increasing magnetic field the increase of the attractive interactions initially is greater than the increase of the electron-electron repulsion, then becomes comparable, and finally the electron-electron repulsion dominates, corresponding to an initial increase and subsequent decrease in binding energy. The electron-electron repulsion for the D^- triplet state is less sensitive to field because the antisymmetric orbital state tends to keep the electrons farther apart on average than the symmetric orbital state of the singlet. When the electron-electron repulsion exceeds the attractive interactions, a D^- ion will dissociate into a neutral donor (D^0) plus an extra electron. In contrast, for a well-center D^- ion, since the peak of the electron probability density along the growth direction coincides with the position of the positive charge, the increase of the attractive interactions is always greater than the increase of the electron-electron repulsion and the kinetic energy with increasing field, which results in a monotonic increase in binding energy.

We have carried out far-infrared magneto-optical studies of two Si-doped $GaAs/Al_{0.3}Ga_{0.7}As$ multiple quantum well

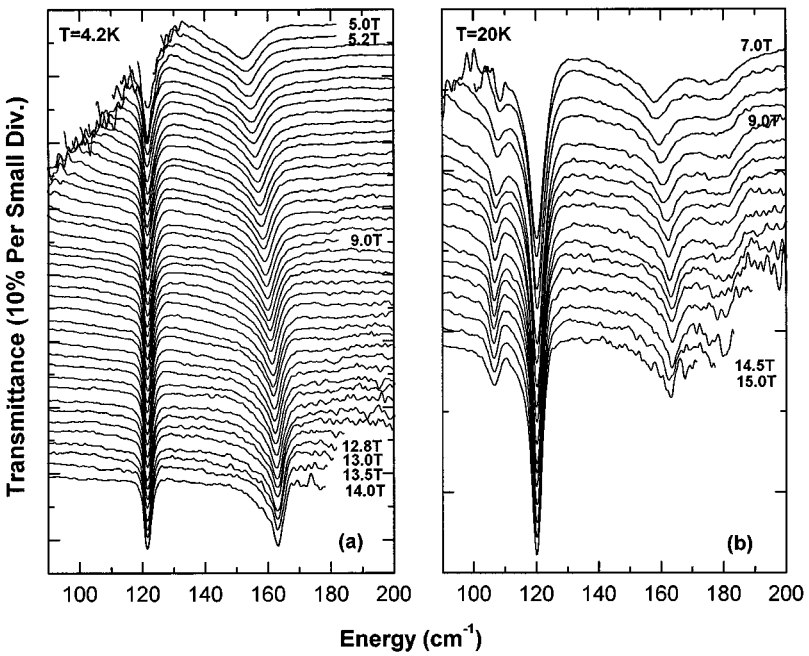

FIG. 1. Magnetotransmission spectra for sample 1. (a) Data taken at 4.2 K in magnetic fields between 5 and 13 T in steps of 0.2 T, and at 13.5 and 14 T; (b) data taken at 20 K in magnetic fields between 7 and 15 T. CR lines for spectra at different fields are aligned to the CR position of the 9-T spectrum to show clearly the behavior of the other features.

(MQW) samples with donor impurities located on and away from the quantum-well centers to compare the different behaviors between well-center and off-well-center D^- ions. As expected, for off-well-center D^- ions the singlet and triplet transition energies (and their binding energies) are found to be smaller and closer to cyclotron resonance (CR), respectively, compared to their well-center counterparts. In striking contrast to the well-center D^- ions, the strength of the off-well-center D^- singlet absorption *decreases* with increasing magnetic field between 5.5 and 15 T due to decreasing population of the ground D^- singlet state. This observation provides direct evidence of the predicted magnetic-field-induced reduction of the singlet binding energy in this system.

Two molecular-beam-epitaxy grown $GaAs/Al_{0.3}Ga_{0.7}As$ MQW structures with 20 periods were investigated. The well barrier width was 200 Å/600 Å. Wide barriers were used to eliminate the possibility of electrons in the wells binding to their parent donor ions in the barriers as neutral donors. Sample 1 is δ-doped with Si donors in both the well centers and the barrier centers at densities of 2×10^{10} cm^{-2} and 3.5×10^{10} cm^{-2}, respectively. Sample 2 is δ-doped at $\frac{3}{4}$ of the distance from the well centers to the top edge of the well in the sample-growth direction at a density of 2×10^{10} cm^{-2} with a barrier-center doping density of 3×10^{10} cm^{-2}. FIR magnetotransmission spectroscopy was carried out with a BOMEM DA-3 Fourier transform infrared spectrometer in conjunction with a 17-T superconducting magnet system, light pipe, condensing cone optics, and a Ge:Ga photoconductive detector. The magnetic field was applied along the sample-growth direction with FIR light propagating along the field (Faraday Geometry).

Magnetotransmission spectra for Sample 1 (well-center D^- ions) are shown in Fig. 1. The major feature at 4.2 K [Fig. 1(a)] is CR; transmission minimum at 121 cm^{-1} at 9 T. In order to show clearly the behavior of the other features, the CR lines for all other spectra at different fields are aligned to the 9-T data. The strong feature at higher energy is the well-center D^- singlet line.[10] Figure 1(b) shows data taken on the same sample at 20 K. The strong feature that appears on the low-frequency side of CR has been identified as the lower-energy triplet transition (T-);[6] elevated temperature is required to populate the ground state of this transition. In addition, evidence of the neutral donor (D^0) $1s$-$2p^+$ line is seen at higher frequencies (transmission minimum at 178 cm^{-1} at 9 T).

Magnetotransmission spectra at 4.2 K for sample 2 are shown in Fig. 2. In addition to the dominant sharp CR feature (transmission minimum at 121 cm^{-1} at 9 T), the strong feature (3) on the high-frequency side of CR increases slightly in separation from CR with increasing magnetic field ($20-25$ cm^{-1} between 5.5 and 15 T). A weak feature (4) appears on the high-frequency side of feature (3), separated by approximately 10 cm^{-1} independent of magnetic field. Another feature (1) is also observed at 4.2 K on the low-frequency side of CR; the increasing separation of this line from CR with increasing magnetic field ($6-13$ cm^{-1}) is apparent.

Temperature-dependence studies[11] indicate that features (1), (3), and (4) are all impurity related; and the binding energies associated with features (1) and (3) are smaller than that for feature (4). From comparison with variational calculations[8] for the off-well-center D^- singlet and the D^0 $1s$-$2p^+$ transitions, features (3) and (4) are assigned to the off-well-center D^- singlet and D^0 $1s$-$2p^+$ transitions, respectively.[11] Furthermore, feature (1) on the low-frequency side of CR has a smaller slope versus field than CR, similar to the lower-energy D^- triplet transition (T-) which appears only at higher-temperatures for well-center D^- ions as indicated in Fig. 1(b). This feature (1) is assigned to the off-well-center D^- triplet (T-) transition. Finally, a weak feature (2) appears between CR and the D^- singlet transition when the magnetic field is larger than 13 T, and increases in strength with increasing magnetic field. This feature is apparently related to barrier neutral donors which had migrated toward the interfaces during sample growth.[11] Since this is not a major focus of this paper, it is not discussed further.

It is worth inspecting Fig. 2 more closely. At low magnetic fields, the D^- singlet line (3) is the dominant impurity-

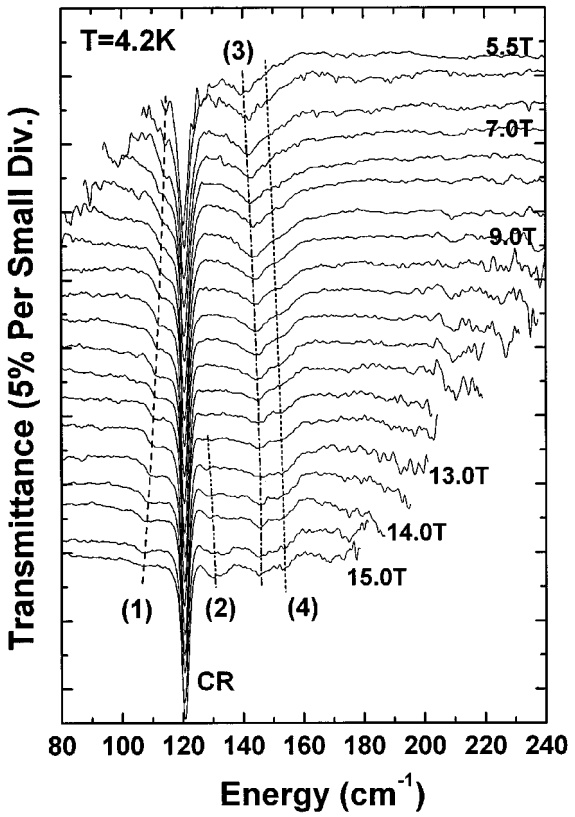

FIG. 2. Magnetotransmission spectra for sample 2 at 4.2 K in magnetic fields between 5.5 and 15 T in steps of 0.5 T. All spectra at different fields are aligned to the 9-T spectrum according to their CR positions.

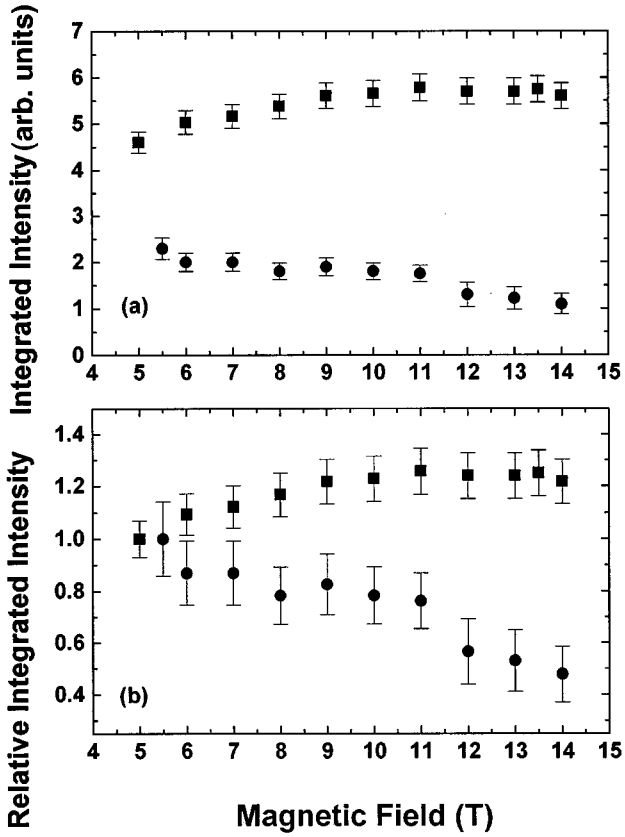

FIG. 3. (a) The integrated intensities (i.e., the areas) of the absorption profiles of D^- singlet transitions as a function of magnetic fields; (b) normalized integrated intensities to that at 5 T. The absorption profiles for the well-center and off-well-center D^- singlet transitions were obtained by taking the logarithm of the corresponding transmittance spectra in Figs. 1(a) and 2, respectively. The solid squares (■) and circles (●) are for the well-center and off-well-center D^- singlet transitions, respectively.

related feature and the D^0 $1s$-$2p^+$ line (4) appears as a very weak shoulder. With increasing field, the strength of the D^- singlet line continuously *decreases* (7% change in transmission at 6 T, 3% at 15 T), while the strength of the neutral donor D^0 $1s$-$2p^+$ line *increases* correspondingly. At high fields, the D^0 $1s$-$2p^+$ line is clearly observable with a strength comparable to the D^- singlet line. We have also carried out similar measurements at higher temperatures (7–20 K), and have observed analogous magnetic-field effects. This behavior is to be contrasted with the strength of the well-center D^- singlet line, which increases slightly with magnetic field [Fig. 1(a)].

In order to obtain a more quantitative comparison between the off-well-center and well-center D^- singlet transitions in Figs. 1(a) and 2, the integrated intensities (i.e., the areas) of the absorption profiles of these transitions, obtained by taking the logarithm of the corresponding transmittance spectra, are plotted as a function of magnetic field in Fig. 3. It is clear that with increasing magnetic field in the range between 5 and 14 T, the integrated intensity of the well-center D^- singlet transition initially increases slightly, and then saturates at high fields, while that of the off-well-center D^- singlet transition monotonically *decreases* by a factor of 2.

The integrated intensity is proportional to the product of the oscillator strength of the associated transition (the product of the square of the absolute value of the dipole-transition matrix element between the initial and final states and the transition frequency) and the number density of occupied initial states, assuming that the final state is unpopulated. In thermal equilibrium, the number density of occupied states depends on the binding energy; i.e., the larger the binding energy, the larger the density. Although there is no theoretical calculation of the oscillator strength of a D^- transition as a function of magnetic field, it is reasonable to assume that the field dependence of the oscillator strength of a D^- singlet transition is qualitatively similar to that of the D^0 $1s$-$2p^+$ transition because two-electron D^- states can be constructed from a linear combination of single-electron (D^0) states modified by the Coulomb repulsion. The oscillator strength of the D^0 $1s$-$2p^+$ transition for a well-center D^0 (150 Å well width) increases slightly with increasing magnetic field in the range between 5 and 10 T (~5%), whereas it remains almost constant for a corresponding edge donor.[12] In addition, the binding energy of the well-center D^- singlet ground state is approximately 2 meV at zero magnetic field,[13,14] considerably larger than k_BT at 4.2 K (~0.35 meV), and it increases with field. Hence, the occupancy of such a ground state should not change significantly with increasing magnetic field at low temperatures. Therefore, the behavior of the integrated intensity of the well-center D^- singlet transition in Fig. 3, an initially slight increase followed by saturation at high fields, is attributed to the field dependence of the oscillator strength of this transition.

On the other hand, the binding energy of the off-well-center D^- singlet ground state for the sample of Fig. 2 is comparable to k_BT at 4.2 K.[8] The carriers are statistically distributed among the ground and excited D^- and D^0 states, and the conduction-band states; the occupancy of a D^- state depends directly on the field-dependent binding energy.[15] The oscillator strength of the D^- singlet transition for a donor located near the edge (which is the case in the present work) should be almost constant in the field range studied. Therefore, the continuous reduction of the integrated intensity of the off-well-center D^- singlet transition represents a monotonic *decrease* of the binding energy of the initial state (ground state) of this transition. This is also supported by the observed *increase* in the strength of the off-well-center D^0 $1s$-$2p^+$ transition, since the dissociation of D^- ions leads directly to the creation of neutral donors according to the ''reaction'' $D^- \rightarrow D^0 + e$.

The binding energy of the off-well-center D^- singlet ground state studied in the present work is predicted to decrease from $\sim$0.9 to $\sim$0.5 meV over the field range 5.5 to 14 T.[8] At 4.2 K, the relative occupancy of the singlet ground state at 14 T to that at 5.5 T, N(14 T)/N(5.5 T), is approximately $\exp[(0.5-0.9)\ \text{meV}/k_BT] \sim 30\%$, in reasonable agreement with the decrease of absorption in Fig. 3(b).

In summary, the off-well-center D^- singlet and low-energy triplet (T-) transitions have been observed. Their transition energies are significantly shifted towards the CR energy compared to well-center D^- transitions as expected. The off-well-center D^- triplet (T-) transition is observed at much lower temperature (4.2 K) than its counterpart in well-center doped samples, which is seen clearly only at elevated temperature (20–25 K). In striking contrast to the well-center D^- ions, increasing magnetic field clearly *reduces* the singlet binding energy of off-well-center D^- ions. This unusual magnetic-field-induced unbinding of the off-well-center D^- state is a direct result of magnetic-field tuning of the competition between the electron-electron repulsion and the attractive interactions of the two electrons with the positively charged impurity center. Our experimental results provide evidence for the predicted magnetic-field-induced unbinding of shallow impurity states and are in good agreement with recent theoretical calculations.[8] The off-well-center D^- ions form a unique system to study electron-electron correlations in high magnetic fields.

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