Evidence for a Stable Excitonic Ground State in a Spatially Separated Electron-Hole System

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Far-infrared magnetospectroscopy on $InAs/Al_xGa_{1-x}Sb$ quantum-well structures has revealed new transitions (X lines) when sufficient holes (in $Al_xGa_{1-x}Sb$) coexist with electrons (in InAs) *and* the magnetic field is high enough. The electron-related X line is about 3 meV above electron cyclotron resonance, has roughly the same slope vs magnetic field, and increases in intensity with increasing hole density or decreasing temperature. This line is assigned to an internal excitonic transition and provides evidence for the existence of an excitonic ground state.

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The exciton is a fundamental excitation in intrinsic semiconductors in which a photoexcited electron and hole are bound together by their Coulomb attraction. For bulk semiconductors at low temperature Bose-Einstein condensation and superfluidity of excitons [1] and Fermi-Dirac condensation and the metallic liquid phase [2] have been discussed for high densities of electrons and holes. For narrow-gap semiconductors or semimetals, Mott, Knox, and others have discussed an excitonic insulator (EI) phase near the semimetal-semiconductor transition (SST), in which all electrons and holes are paired to form a charge neutral exciton gas when the binding energy of the exciton, E_X, is larger than the absolute value of the energy gap, $|E_G|$ [3]. Under this condition, the EI phase is the ground state of the system, and excitons can be formed spontaneously at low temperature without photoexcitation. However, the EI phase has not been conclusively observed [4].

Recently, spatially separated electron-hole (e-h) systems have been proposed to study the EI phase [5,6]. Although the electrons and holes are spatially separated in one direction, calculations have shown that E_X can be large compared with the thermal energy at 4.2 K, and E_X increases as the barrier width decreases [5,6]. The $InAs/Al_xGa_{1-x}Sb$ quantum well (QW) structure represents a limiting case with zero barrier width. In addition, the band alignment between the two materials (thus the effective "energy gap", E_G^*) can be tuned by changing the Al composition, x. For $x = 1$, the InAs conduction band is ~300 meV higher than the AlSb valence band, a staggered type-II semiconducting heterostructure ($E_G^* > 0$). When $x = 0$, the InAs conduction band is lower in energy than the GaSb valence band by about 150 meV, a broken-gap type-II heterostructure which is semimetallic with a *negative* E_G^* (electrons in InAs and holes in GaSb). The SST takes place as E_G^* vanishes near $x = 0.3$ at zero magnetic field. In the semimetallic phase ($E_G^* < 0$), the envelope function overlap between the electrons (InAs) and the highest lying (heavy) holes (AlGaSb) at the interface is very small due to the symmetry mismatch of the predominant band-edge functions in the adjacent materials.

We report a far-infrared (FIR) magneto-optical study of a set of $InAs/Al_xGa_{1-x}Sb$ QW structures in which the Al composition varies from $x = 0.1$ to 1.0. We have observed new spectroscopic features (X lines) which appear only when holes and electrons coexist *and* the magnetic field is high enough. The magnetic field and temperature dependences of the X lines are consistent with assigning them to intraexcitonic transitions of excitons formed via Coulomb attraction between spatially separated e-h pairs and stabilized by the magnetic field. These experimental observations provide strong evidence for the existence of an excitonic ground state for a spatially separated two-dimensional (2D) e-h system.

The six samples used in this experiment were grown by molecular beam epitaxy under the same growth conditions. Only the Al compositions of the $Al_xGa_{1-x}Sb$ barrier layers were varied ($x = 0.1$, 0.2, 0.4, 0.5, 0.8, and 1.0). The nominal structure of all samples is a 3 μm $Al_xGa_{1-x}Sb$ buffer layer grown on a semi-insulating GaAs substrate, a 15 nm InAs single QW, a 15 nm $Al_xGa_{1-x}Sb$ top barrier, and a 10 nm GaSb cap layer. The samples are all n type as grown with excess electrons trapped in the InAs well [7]. The 2D electron densities at 4.2 K are 6.2 (5.5) $\times 10^{11}$, 6.4 (5.8) $\times 10^{11}$, 6.4 (5.4) $\times 10^{11}$, 8.5 (6.1) $\times 10^{11}$, 8.2 (5.0) $\times 10^{11}$, and 9.5 (6.0) $\times 10^{11}$ cm^{-2} for the samples in the sequence listed above. Numbers in parentheses are densities after illumination, which reduces the electron density (thus the Fermi energy) due to the negative persistent photoeffect [7]. The corresponding mobilities at 77 K are 1.1×10^5, 1.7×10^5, 0.9×10^5, 2.2×10^5, 0.5×10^5, and 0.3×10^5 cm^2/V s. The 2D

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hole densities at zero field after illumination were estimated to be $\sim(1-2)\times 10^{11}$ cm^{-2} for the $x = 0.1$ sample from high temperature cyclotron resonance (CR) intensities, and $\sim 0.3\times 10^{11}$ cm^{-2} for the $x = 0.2$ sample from magnetotransport measurements [8]. FIR magnetotransmission spectra were obtained with Fourier transform spectrometers in conjunction with superconducting magnets (in Buffalo) or a Bitter-type magnet (at FBNML). A red light-emitting diode (LED) was mounted *in situ* to provide illumination to the samples.

Transmission spectra (normalized by ratioing to zero-field spectra) for three samples ($x = 0.5$, 0.2 and 0.1) at 4 T are shown in Fig. 1. The upper (lower) traces in each panel are spectra taken before (after) LED illumination. For the sample with $x = 0.5$ ($E_G^* > 0$), CR of 2D electrons in the InAs well is the only absorption line in the spectral region of interest. After LED illumination, the CR integrated intensity is reduced by about 28%, while the transmission minimum shifts to higher energy (a lighter effective mass). Both the decrease in intensity and the position shift are consistent with the single particle picture; the Fermi energy (electron density) is lowered by LED illumination, and the CR mass is reduced due to the nonparabolicity of the InAs conduction band [9]. All the other semiconducting samples ($x = 0.4$, 0.8, and 1.0) showed qualitatively the same behavior. For the sample with $x = 0.2$, the valence band edge of $Al_{0.2}Ga_{0.8}Sb$ is slightly higher than the Fermi energy of confined electrons at zero field; hence, a small number of holes should

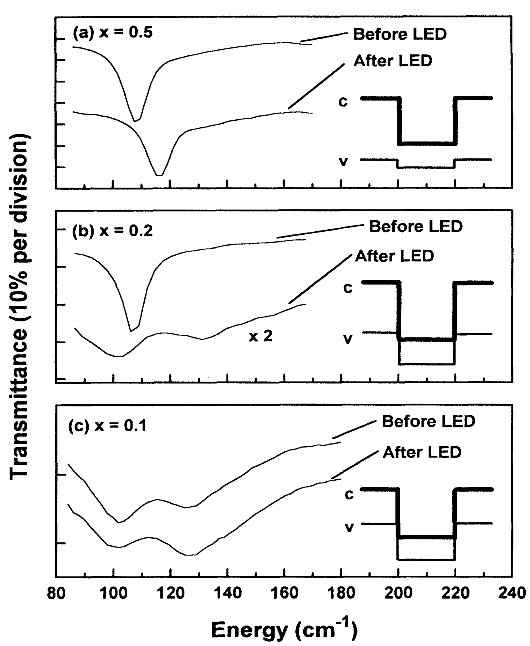

FIG. 1. Transmission spectra at 4 T before and after exposure to an *in situ* red LED: (a) $x = 0.5$, (b) $x = 0.2$, and (c) $x = 0.1$. Insets show schematically the ideal (charge transfer or band bending not included) conduction (thick lines) and valence (thin lines) band lineups for the three samples.

exist in the $Al_{0.2}Ga_{0.8}Sb$ layer(s). The number of holes decreases with increasing magnetic field and vanishes at about 4–5 T in the absence of LED illumination; i.e., a magnetic-field-induced SST occurs [8]. After LED illumination, the Fermi energy is lower; hence, the transition field moves up to about 7–8 T (see Ref. [8]). Before LED illumination, the FIR spectra for this sample [Fig. 1(b)] show a single sharp CR line over the whole field and energy ranges of the experiment. Illumination results in a dramatic change in the spectra when the field is larger than ~4 T. An additional feature, the e-X line, appears 30 cm^{-1} above CR. The intensity of the CR line decreases dramatically while it shifts to *lower* energy and broadens slightly. The enhanced CR mass after illumination is opposite to that of the semiconducting samples and cannot be explained by the usual single-particle band nonparabolicity. For the sample with $x = 0.1$ [Fig. 1(c)], the e-X line is observable at fields above ~1 T, both before and after LED illumination, with increased intensity after illumination. Both lines shifted down in energy when we tilted the magnetic field from the growth axis, indicating that they are 2D in nature.

In Fig. 2 we plot the transition energy (a) and the absorption amplitude (b) as a function of magnetic field up to 7 T for both CR and the X lines for the sample with $x = 0.1$ after illumination. Both CR and e-X line show an overall linear dependence on field with essentially the same average slope; at fields near 3, 4, and 6 T (corresponding to $\nu = 8$, 6, and 4, respectively, where $\nu = n_s hc/eB$ is the Landau level filling factor), the transition energies show slope discontinuities [10]. The equivalence of the slopes shows that the e-X line is related to 2D electrons in the InAs QW. The amplitude of the e-X line increases rapidly with magnetic field at the expense of CR in the low-field region ($B < 3$ T) [see Fig. 2(b)], and it oscillates at higher fields with maxima coincident with the minima in the CR intensity close to even ν [10]. Above 3 T another low frequency line is observed (the h-X line) with an average slope corresponding to a mass of $0.1m_0$ as shown in Fig. 2(a).

The temperature dependence of the e-X line has been studied at several magnetic fields. At 5 T and 4.2 K, the e-X line dominates the spectrum, as shown in Fig. 3. With increasing temperature, CR increases in intensity rapidly while the e-X line decreases. Above 20 K CR becomes dominant, and above 50 K the e-X line is a weak shoulder on the high energy side of CR. The spectra can be fitted very well by two Lorentzians, as indicated in the figure. The clear decrease in the intensity of the e-X line at higher temperatures suggests that it is associated with a bound state. At other magnetic fields, the general temperature dependence is the same, but details are sometimes very different (e.g., the positions shift with temperature), especially at fields close to $\nu = 8$, 6, and 4. Similar behavior is observed for the h-X line; as temperature is increased the line gradually shifts to lower frequency toward hole CR with light in-plane mass.

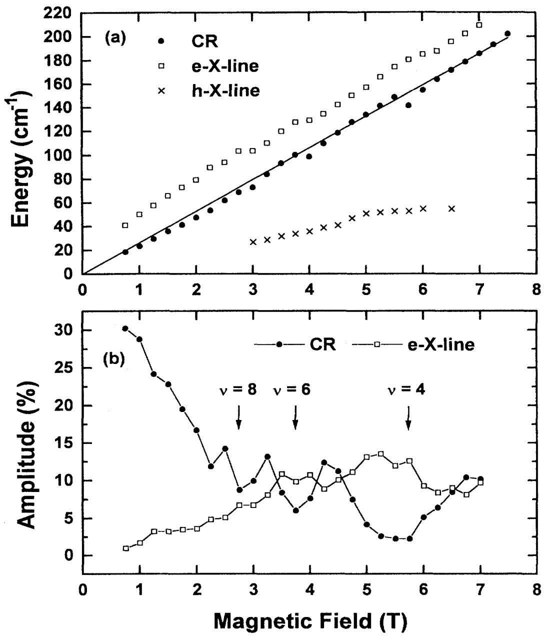

FIG. 2. Results for a sample with $x = 0.1$ after exposure to a red LED. (a) Energy vs magnetic field for the three lines at 4.2 K. The straight line has a slope corresponding to $m^* = 0.0352m_0$. (b) Plot of percent change in transmittance at peak of the two lines vs magnetic field.

We have considered and *excluded* several possibilities for the origin of the e-X line: (1) Spin-resolved CR, which has been observed in InAs/AlSb QW structures [11], exhibits two (or more) peaks at certain fields, but the energy separation between the two lines is much smaller than the CR-X separation. In spin-resolved CR a low-energy line should disappear and be replaced by a high-energy line at integer (even *or* odd) filling factors [11], clearly different from the behavior of the e-X line. In fact, spin-resolved CR's have been observed for the samples at high fields (three lines were observed at fields above 13 T for the sample with $x = 0.1$, and two lines with about $5-6$ cm^{-1} separation for the samples with $x = 0.4$ and 0.5 at 6–7 T); they are not related to the e-X line. (2) An additional CR of electrons in the second subband, as reported in previous work on InAs/GaSb multiheterostructures [12], yields two peaks, but the separation between the two lines should increase with increasing magnetic field because they have different masses. Also, from the sample dimensions and densities, it is clear that a second subband cannot be populated in our samples [13]. (3) Residual impurities binding electrons in the InAs wells can yield an absorption line above CR. However, with such a large density of electrons in the wells, negative donor ions (D^-) are the only possibility, which should exhibit smaller transition energies (relative to CR) with slightly larger slope versus field and a much more rapid loss of intensity with temperature because of very small binding energy [14]. More importantly, the e-X line is observed only *in the samples that are semimetallic* (and in the case of the $x = 0.2$ sample only *after* illumination which creates holes). There is no evidence of the e-X line in semiconducting samples ($x = 0.4$, 0.5, 0.8, and 1.0), although background impurity density should be similar for *all the samples.* In addition, the observation of the h-X line only in the strongly semimetallic sample is completely inconsistent with shallow donor impurities associated with InAs. (4) Quasi-2D magnetoplasmons should appear at energies above CR and have a field dependence nearly equal to CR in high magnetic fields. However, coupling of FIR light to the plasmon modes at finite wave vector $\boldsymbol{k}$ requires a well-defined periodic lateral modulation in the Faraday geometry, which we believe to be extremely unlikely to result from say random impurity potentials. The strongest argument against this interpretation is the fact that the e-X line does not appear in *any* of the semiconducting samples, in particular in the sample with $x = 0.5$, which has the largest electron density (hence the largest plasmon frequency) as well as the highest mobility. In addition, the fact that the e-X line appears only after LED illumination (lower electron density) for the sample with $x = 0.2$ also argues against the magnetoplasmon interpretation. (5) CR of localized electrons can have the same qualitative magnetic field dependence [15], but the random fluctuating potential, which causes electron localization, is important only when the density (filling factor) is very small ($\nu < 2$), outside the range of this experiment.

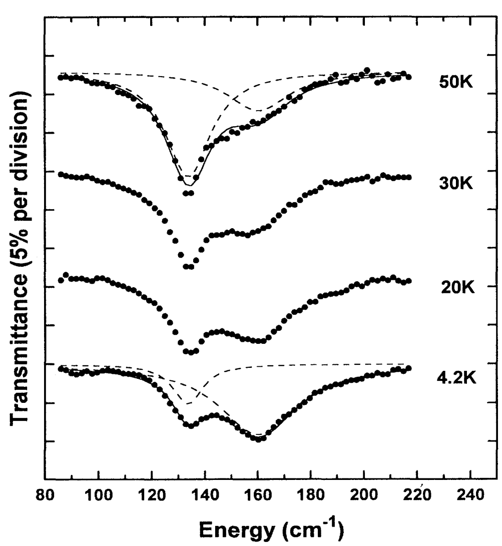

FIG. 3. Temperature dependence of the two lines for a sample with $x = 0.1$ after exposure to the LED. Ratios of transmitted intensity at the indicated temperature and $B = 5$ T to a background spectrum (at the same temperature and $B = 0$) are shown by the dotted curves. Individual Lorentzian lines fitted to the data (dashed lines); overall fit (solid lines).

On the other hand, all of the experimental observations are qualitatively consistent with the assignment of the e-X and h-X lines to intraexciton transitions ($1s$-$2p_+$-like for electrons and holes, respectively). The excitons are formed through the Coulomb attraction between the spatially separated electrons and holes, and they are stabilized by applying a magnetic field perpendicular to the plane. For semiconducting samples ($x = 0.4$), neither of the X lines is observed, since there are no holes. For the sample with $x = 0.2$ before illumination, although there *are* holes at low fields ($B < 4$ T), the density is not large enough to observe the excitonic transitions, and increasing magnetic field serves to decrease the hole density. After illumination, the Fermi level moves down, the density of holes increases, persisting to high magnetic fields [8], and the e-X line is observable over the field region 4 to 7 T. For the sample with $x = 0.1$, holes exist over the whole field region, and the hole density for this sample is larger than that of the sample with $x = 0.2$ due to larger band overlap. Therefore, both the e-X and h-X line are stronger. The binding energy of the exciton can be estimated to be roughly 4 meV from the CR-X separation in the hydrogenic model. This is consistent with the calculated binding energy for similar systems [5,6], but much larger (a factor of 3–4) than that for InAs/GaSb single QW's [16]. This is probably because the calculation in Ref. [16] does not take into account the confinement of holes in the growth direction which should increase the binding energy significantly. Field should also increase the binding energy drastically [5,17], and more detailed calculations including these effects are needed to compare with our data. The exciton can be thermally ionized at high temperature, decreasing the e-X and h-X line intensities, and increasing the free electron (hole) CR intensities. The slope of the e-X and h-X line can be understood from simple considerations. For excitons the use of the reduced mass is correct in the low-field limit; however, in the high field limit appropriate to the present situation ($\gamma = \hbar\omega_c/2\mathrm{Ry}^* = 1$ corresponds to 1.4 T, where Ry* is the electron effective Rydberg), it can be shown [18] that the internal transition near electron (hole) cyclotron resonance is determined by the electron (in-plane hole) effective mass.

A few features still remain unexplained. First, excess electrons should play an important role just as in the D^0 and D^- situations [14]; the anomalous behavior in the transition energy of the X line at even filling factors is evidence for this. A simple static screening model predicts a significant reduction in the binding energy due to excess electrons; however, recent results on "screening" of shallow impurities in QW [14,19] show that excess electrons can cause a blue (rather than red) shift in the transition energy. Second, the previous FIR experiment on InAs/GaSb QW's [20] did not reveal an analogous X line, although more holes presumably exist in GaSb in comparison with our samples. This, aside from the sample quality and differences in surface Fermi-level pinning, might be due to the fact that the electron-electron and hole-hole interactions dominate the electron-hole interaction when the densities of carriers are high, and the ground state of such a two-carrier many-body system should not be the excitonic phase at higher densities.

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