Shakeup processes in the recombination spectra of negatively charged excitons

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(Received 2 January 1996)

We report on shakeup processes in the luminescence spectra of a negatively charged exciton (X^-) at moderate magnetic fields. These processes manifest themselves as a series of low-energy satellite peaks emerging from the negatively charged exciton line. We analyze the dependence of the X^- energy on magnetic field. We conclude that at magnetic fields above ~ 1 T the X^- structure can be viewed as an electron at the lowest Landau level bound to an exciton. [S0163-1829(96)07520-0]

Negatively charged excitons (X^-) consisting of two electrons bound to a hole were observed recently in the optical spectra of semiconductor quantum wells.[1–6] This bound complex appears as an absorption or emission line below the neutral exciton line in a number of systems: modulation-doped quantum wells,[1–3,5] resonant tunneling diodes,[4] and mixed-type quantum wells.[6] Common to all these systems is the presence of a reservoir of electrons in the well, which can bind to the photoexcited electron-hole pairs and form the X^-. This reservoir is provided by donors in modulation-doped structures, by current injection in resonant tunneling devices and by electron-hole spatial separation in mixed-type crystals. Very recent observations have demonstrated that by p-type modulation doping or by current injection of holes, one can create its positive counterpart, the positively charged exciton.[7–9]

A particularly convenient system for studying the X^- is the gated two-dimensional electron gas (2DEG) in a quantum well. In this system the electron density could be changed in a controlled and reversible manner by applying a gate voltage. Furthermore, by conducting transport measurements we can accurately determine the 2DEG density and its mobility at any applied gate voltage. Studying the correlation between the evolution of the optical spectra and the transport properties in a number of samples as the electron density in the well is reduced we have shown the appearance of the neutral and charged excitons lines at the metal-insulator transition.[2] A photoexcited electron hole pair in these conditions could either form a neutral exciton or bind with a localized electron to form an X^-. The localization of the electrons in the potential fluctuations of the remote ionized donors makes the 2DEG inefficient in screening the X^-.

The energy spectrum of X^- in a magnetic field was the focus of a few recent works. It was shown that the energy position of the X^- exhibits a diamagnetic shift, which is very similar to that of the exciton.[4] At strong magnetic fields, when the magnetic length becomes smaller than the X^- diameter, a new line, the X_t^-, is observed.[7,8] This line is attributed to the negatively charged exciton where the two electrons are arranged in a triplet state. Finally, it was shown that the Zeeman splitting of the X^- is different from that of the neutral exciton, but is very similar to that of a 2DEG.[7]

In this work we wish to extend these studies and investigate shakeup (SU) processes in the recombination spectrum of the X^-. In these processes the recombination of an electron and a hole in X^- is accompanied by ejecting the second electron to some high-energy state. At a finite magnetic field this energy state is a higher Landau level (LL). Consequently, the process gives rise to a fan of discrete lines, each associated with a different LL as the final state for the second electron. In a metallic 2DEG such recombination lines were first observed by Nash *et al.* in the magneto-photoluminescence of $In_xGa_{1-x}As/InP$ modulation-doped quantum wells.[10] The SU lines were seen as satellite peaks at energies that are close to an integer number times $\hbar\omega_c$ below the main recombination line. Very recently, van der Meulen *et al.* have studied the dependence of the intensity of these peaks on the magnetic field and found that they are maximized at even-integer filling factors ν.[11] Some earlier studies of SU processes in GaAs had been reported by Sooryakumar *et al.*, who found wave-vector nonconserving photoluminescence and related it to SU processes.[12] Potemski *et al.* have observed emission at an energy higher than the exciting light, and related it to an Auger-like excitation of electrons to a higher LL.[13]

In the present work we study the low-energy recombination spectrum of the X^- line in a magnetic field and identify a series of SU peaks. We find that the energy separations between these peaks roughly follow $\hbar\omega_c$ dependence on magnetic field B, where $\omega_c = eB/m_e c$ and m_e is the electron effective mass. The second part of the paper is devoted to studying the low-energy tail of the X^- emission spectrum at zero magnetic field. We conclude that this tail is not related to SU processes, but is rather due to localization of the recombining electrons and holes.

We have studied two single-sided modulation-doped quantum-well samples. The samples structure consists of a buffer superlattice, a 200-Å quantum well, an undoped spacer layer, a Si δ-doped region, and a cap layer (an additional uniform doping is introduced near the surface). The only parameter that was changed was the spacer width: it was nominally 1500 Å in the first sample and 500 Å in the second. The electron densities at zero gate voltage after illumination are $\sim 7\times 10^{10}$ and $\sim 2\times 10^{11}$ e/cm^2, respectively. Samples mobility was about 10^6 $\mathrm{cm}^2/\mathrm{V\,sec}$.

The samples were processed to form 1.2×1.2-mm mesas. Ohmic contacts were formed into the 2DEG and a semi-transparent gate was evaporated. The electron density was varied by applying voltage to the gate with respect to the contacts. The incident laser power density was about 100

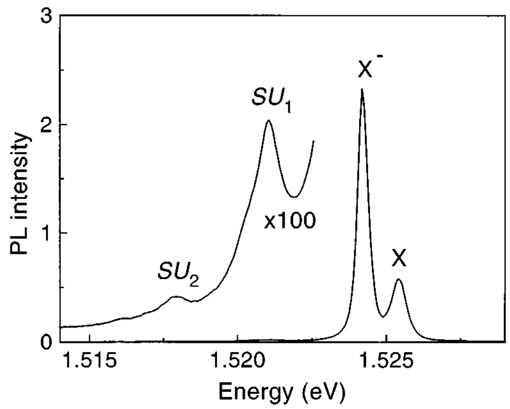

FIG. 1. The PL spectra of the 1500-Å spacer sample in the insulating state at 2 T. The low-energy region is multiplied by a factor of 100. X, X^-, SU_1, and SU_2 are visible.

$\mu W/cm^2$ at 632 nm. The photon energy is lower than the barriers' energy gap, thus avoiding space-charge buildup by carriers, which are photoexcited in the barriers.[14] The measurements were performed at $T=4.2$ K using a magnetic field in the range of 0–9 T. Light was coupled in and out using optical fibers and the detection of the photoluminescence (PL) was done with a cooled charge-coupled device camera.

Figure 1 displays the PL spectra of a 1500-Å spacer sample at a gate voltage of $V_g=-3.1$ V and a magnetic field of 2 T. This gate voltage corresponds to an insulating state of the 2DEG, and one can clearly see the neutral (X) and negatively charged (X^-) excitons. Looking more closely at the low-energy tail of the X^- we can observe two satellite peaks, SU_1 and SU_2, below the X^- line. Following the energy position of these peaks we observe a monotonic shift to lower energies with increasing magnetic field. In Fig. 2 we show the energy positions of the SU peaks as a function of magnetic field for the 500-Å spacer sample. One can see that the shift in energy is linear with magnetic field, and the slope of the SU_2 line is approximately twice that of the SU_1. In fact, in both samples one could observe a larger number of peaks at about 1 T: up to 5 satellite peaks in the 1500-Å spacer sample and 4 in the 500-Å spacer sample. The energy

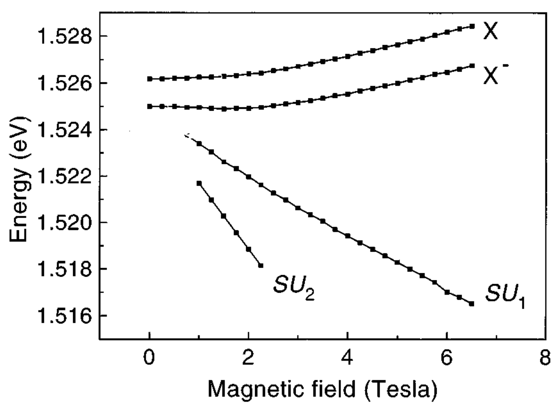

FIG. 2. The X, X^-, SU_1, and SU_2 peaks energies as a function of magnetic field for the 500-Å spacer sample in the insulating state.

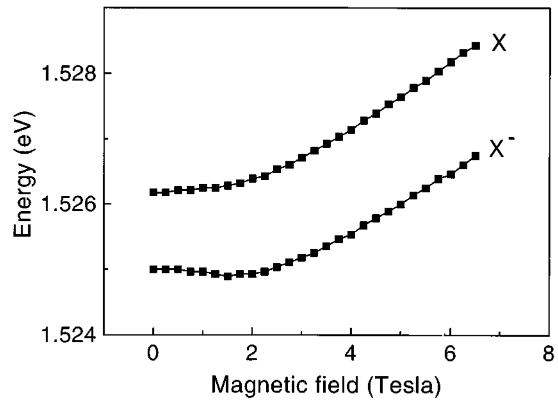

FIG. 3. The X and X^- peak energies as a function of magnetic field for the 500-Å spacer sample in the insulating state.

difference between SU_1 and SU_2 lines and between X^- and SU_1 lines in both samples is ≈ 1.55 meV/T. This slope is slightly smaller than the expected $\hbar\omega_c$: taking the mass to be the GaAs electron effective mass ($m_e=0.067m_0$) one would get 1.73 meV/T. The energy states we are dealing with are close to the bottom of the conduction band. Thus the band nonparabolicity is an improbable explanation for this 10% discrepancy. A possible explanation is that the effective mass depends on the penetration of the wave function to the $Al_xGa_{1-x}As$ layer, where m_e should be $\approx 0.1m_0$.

The appearance of multiple peaks, the linear dependence of their energies on the magnetic field, and the fact that these energies are close to $n\hbar\omega_c$ could be taken as proof that the satellites are due to SU processes: the recombination of one of the two electrons in X^- with the hole, leaving the second electron on one of the higher LL's. This assignment of the peaks is supported by the dependence of their intensity on magnetic field. We observe that the intensity of the SU peaks decreases with increasing magnetic field and eventually disappears above 5 T. This behavior can be explained by a simple wave function overlap argument: at high magnetic fields, the main contribution to the wave function of an electron in X^- is made by the lowest LL states. The overlap of this wave function with the higher LL decreases with increasing field and the SU process becomes less probable.

In Fig. 3 we plot the energy position of the exciton and the X^- as a function of magnetic field. The X^- line corresponds to a recombination of one of the electrons with a hole, while the other electron is left on the lowest LL. It was argued that this process should result in a negative slope of the X^- energy with increasing magnetic field.[15] The reason for this is the $\frac{1}{2}\hbar\omega_c$ energy of the electron, which is left after the recombination process. It is evident from Fig. 3 that the X^- recombination energy is indeed decreasing with increasing field at the field range $0<B<2$ T, but the decrease is much smaller than $\frac{1}{2}\hbar\omega_c$. A similar behavior was observed in other works on GaAs/$Al_xGa_{1-x}As$ and CdTe/$Cd_xZn_{1-x}Te$ quantum wells.[8,16] This small negative energy shift was not observed by Buhmann *et al.*,[4] possibly due to the large width of their PL lines. Kheng *et al.*[16] attributed the small value of the shift to the strong localization of X^- by the random potential fluctuations. They argued that the electron, which is left after the recombination process, is localized and therefore does not acquire the $\frac{1}{2}\hbar\omega_c$ energy. The

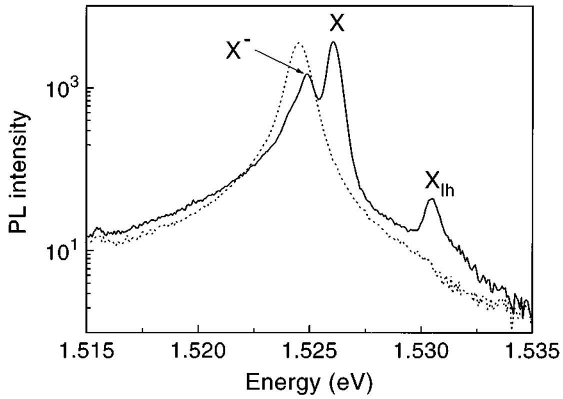

FIG. 4. The PL spectra of the 500-Å spacer sample close to the metal-insulator transition at zero magnetic field. Dashed: the gate voltage is -1.45 V (metallic state), solid: the gate voltage is -1.55 V (insulating state). Note the low-energy tail that appears in the insulating state. X_{lh} is the light-hole exciton transition.

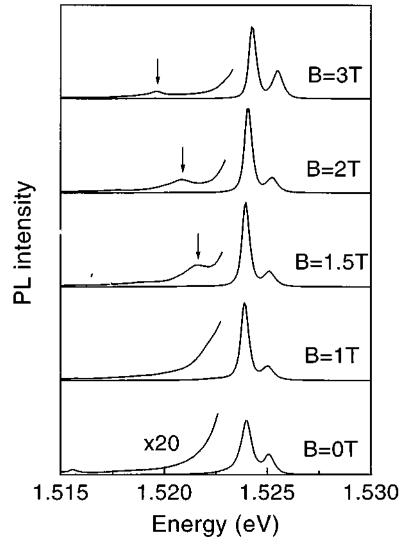

FIG. 5. Series of PL spectra of the 1500-Å spacer sample in the insulating state at 0, 1, 1.5, 2, and 3 T (from bottom to top). The low-energy regions are multiplied by a factor of 20. SU_1 peak is marked by arrows.

observed behavior of the SU peaks, however, indicates that in our samples localization has a relatively small effect on the energy spectrum of the electron left after recombination. Furthermore, if the localization would have played a significant role, one could expect sample dependence of the X^- energy slope. However, we find that the difference between the exciton and the X^- recombination energies in magnetic field is very similar in different samples with different spacer widths and doping concentrations.

This weak magnetic field dependence of the X^- line calls for a reexamination of the basic assumptions concerning the description of X^- in a magnetic field. The expectation that the X^- line would exhibit $-\frac{1}{2}\hbar\omega_c$ behavior is actually based on the decoupling of the internal motion within X^- from the motion of its center of mass.[15] In the presence of the magnetic field this decoupling is only approximate. The internal motion should result in a diamagnetic shift, a quadratic dependence of the energy on the magnetic field. The center-of-mass motion should give rise to a fan of LL with a very small spacing corresponding to a large X^- mass, $m_{X^-} = 2m_e+m_h$. Both contributions are small compared to the $\frac{1}{2}\hbar\omega_c$ energy of the electron left after the recombination. Accordingly, the latter quantity should determine the observed behavior of the X^- recombination energy in magnetic field. However, our results indicate that this decoupling of the internal and the center-of-mass motions stops being valid already at very small fields, not exceeding 1 T. Rather, at $B \gtrsim 1$ the structure of X^- can be approximated by an electron at the lowest LL bound to an exciton. In the process giving rise to the X^- line this electron is left on the lowest LL after recombination. Consequently, the difference between the exciton and the X^- recombination energies is only slightly field dependent. It seems reasonable that the crossover from a zero-field behavior, of two electrons bound to a hole, to the high-field behavior, of an electron bound to an exciton, occurs at $B\sim 1$ T. Approximately at this field the cyclotron energy becomes comparable with the binding energy of the second electron in X^-.

We now wish to turn to a discussion of the low-energy tail of the PL spectra at zero magnetic field. Figure 4 displays the PL spectrum of the 500-Å spacer sample at a gate voltage $V_g = -1.45$ V, where the 2DEG is in a metallic state, and at $V_g = -1.55$ V, where it is insulating. The metal-insulator transition in this sample occurs at about -1.5 V and one can clearly observe the changes in the PL spectrum that accompany this transition: from a single peak at the metallic state (dashed line) to an exciton-X^- doublet at the insulating state (solid line).[2] Examining the low-energy part of the spectra one can clearly see a tail, which extends up to ~ 10 meV below the main peaks. This tail appears in both samples and will be the subject of the following part of the paper.

The question that immediately arises is the relation between this low-energy tail at zero magnetic field and the SU peaks, which appear at $B>0$. Figure 5 displays the evolution of the low-energy tail as the magnetic field is increased from 0 to 3 T for the 1500-Å spacer sample. It is seen that as the magnetic field is increased the tail disappears and is replaced by the SU peaks. In fact, the amplitude of the SU peaks at $B>0$ is similar to that of the zero-field tail, at the corresponding energy. This gradual evolution of the low-energy tail into the set of discrete SU peaks makes it plausible to suggest that the tail originates from SU processes, which occur at zero magnetic field. However, as we show in the following, the dependence of the tail on the incident laser power density and on the temperature is inconsistent with this suggestion.

We first examine the dependence of the tail on the incident laser power density. We vary the incident laser power over five orders of magnitude, from ~ 100 nW/cm^2 to ~ 10 mW/cm^2. As expected, we find that the exciton and X^- lines grow linearly with intensity. On the other hand, the tail grows slower than linearly at intensities larger than ~ 10 μW/cm^2. Moreover, the lower-energy parts of the tail saturate before the high-energy parts.

Next, we study the temperature dependence of the PL tail in the range 4–15 K. It is well known that the X^- line decreases with increasing temperature, due to a thermal ionization into an exciton, and the ratio of the two follows $e^{\Delta/T}$, where Δ is the X^- binding energy.[1,2,4] However, we find that the PL tail at energies larger than ~ 2 meV below X^- is almost unaffected by the temperature change.

These observations indicate that PL tail behavior is different from that of X^-. This result is inconsistent with the assumption that the low-energy PL tail originates in recombination of the same charged excitons accompanied by SU processes. Indeed, if it would be so, the PL tail would follow X^- temperature and incident power dependence.

Alternatively, one can explain the low-energy tail as originating from recombination at some low-energy sites. The sublinear dependence of the PL tail on energy then could be explained as being due to the saturation of these sites. In this case the tail originates from both exciton and X^--like states and it should not follow the temperature dependence of X^-. In this explanation, the disappearance of the tail with increasing magnetic field is due to a decrease of the electron-hole pair diffusivity, giving rise to a smaller probability of reaching a low-energy site. The abrupt appearance of these recombination channels at the metal-insulator transition reflects the rapid growth of the random potential fluctuations.[2] In this situation, rare low-energy configurations of recombining electrons and holes become more probable. This gives rise to a rapid appearance of the exciton-X^- tail (Fig. 4).

In conclusion, we have observed shakeup processes in the negatively charged exciton photoluminescence at finite magnetic field. The analysis of the energy of X^- in magnetic field in combination with SU spectra allows us to conclude that at $B \gtrsim 1$ T the structure of X^- should be approximated by an electron bound to exciton. Also we studied the long low-energy tail of an exciton–negatively charged exciton PL doublet at zero magnetic field. We attributed it to a recombination of localized electrons and holes from rare deep sites rather than to shakeup processes.

This work was supported by the Israeli Academy of Science.

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