Oscillator strengths of dark charged excitons at low electron filling factors

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By direct absorption spectroscopy and comparison to photoluminescence (PL), we investigate negatively charged excitons in dilute two-dimensional electron systems at temperatures down to $T = 40$ mK in the regime of the fractional quantum Hall effect. At very low temperatures, for filling factor $\nu < 1/3$, an additional excitation appears in the PL spectrum, between the well-known singlet and triplet excitons. The observation of a similar excitation by PL was reported very recently [G. Yusa *et al.*, Phys. Rev. Lett. **87**, 216402 (2001)], and the excitation was assigned, in spite of the PL intensity similar to that of the neutral exciton, to be due to a dark triplet exciton. By comparing PL and direct absorption spectra in optically thin samples at $T < 100$ mK, we can identify the new excitation indeed as a "dark" mode, since we find that the oscillator strength is much smaller than those of the "bright" modes.

The fractional quantum Hall effect (FQHE), which is a direct manifestation of many-particle interactions in a two-dimensional electron system (2DES), has attracted great and sustaining research interest since its discovery.[1] The past decade has shown that, in addition to transport experiments, optical spectroscopies, especially photoluminescence (PL) spectroscopy, can provide valuable further information about the ground state properties of the systems. In fact, this method has in recent years been applied to study an interacting 2DES in the regime of the FQHE,[2–5] where a variety of anomalies in both, the energetic positions and the intensities of the PL lines, were found. The striking difference in optical experiments, compared to electron transport, is the presence of photo-excited holes. Though it was previously shown that in symmetric systems in the high magnetic field limit the electron-electron and electron-hole interactions cancel exactly due to a hidden symmetry,[6–9] for most of the real systems this symmetry is violated.

In order to systematically understand the interactions between electrons and photo-excited holes in modulation-doped systems, it seems natural to start with zero density of the 2DES and gradually increase the number of free electrons. In fact, it was found that in dilute 2DES the Coulomb attraction between electrons and holes leads to formation of negatively charged excitons, consisting of two electrons and one hole.[10–12] It is now commonly accepted that at zero magnetic field the two electrons form a spin singlet state (total spin $S = 0$). At finite magnetic field, an additional bound state exists, where the electrons form a spin triplet ($S = 1$).[13,14] It was calculated that at high magnetic fields the triplet exciton (X_t^-) forms the ground state of the system. Magnetic fields of 30–40 T were predicted for the singlet triplet crossing for *symmetric* quantum-well systems.[15–17] For some time, experimental attempts to detect this singlet triplet crossing failed. Instead, a saturation of the X_t^- binding energy was found at high fields.[13,14,18,19] This puzzle was recently solved by a theoretical work of A. Wojs *et al.*[20,21] The authors calculated the spectrum of charged excitons by exact diagonalization of finite size systems. It was found[20] that at high magnetic field *two different* triplet excitons exist as bound

states. One of these excitons has a total angular momentum of $\mathbf{L} = 0$, and is therefore called the *bright* triplet (X_{tb}^-). The second triplet exciton is called the *dark* triplet (X_{td}^-) because it cannot decay radiatively ($\mathbf{L} = -1$). The calculations showed that the X_{td}^- becomes the ground state at high fields and the experimentally observed saturation of the binding energy can be attributed to the bright triplet. In a subsequent work, I. Szlufarska *et al.* found that in *asymmetric* quantum wells the singlet triplet crossing can occur at considerably lower fields.[22] Very recently, the observation of an excitation in PL experiments, which was interpreted as a dark triplet exciton, was reported by two different groups.[23,24] The singlet triplet crossing occurred at $B = 40$ T[23,25] and $B = 15$ T.[24] However, there seem to be some discrepancies between these reports, e.g., in Ref. 23 the dark triplet is visible over the whole magnetic field range.

In this Rapid Communication we report results which are concerning the PL experiments consistent with the observation of G. Yusa *et al.*,[24] i.e., (i) we observe a charged exciton at very low temperatures, which is not present in the spectrum at $T = 2$ K, (ii) this exciton occurs for filling factor $\nu < 1/3$ only, i.e., related to the FQHE regime, and, (iii) the PL intensity of this excitation is of similar strength as that of the singlet exciton X_s^-.

At the first sight it is intriguing that a *dark* mode should appear in the PL spectrum with almost equal strengths as the *bright* modes, since its oscillator strength should by definition be exactly zero. However, it is well known that the PL intensity and linewidth might not directly reflect the oscillator strength of a transition, which is proportional to $1/\tau_X$, where τ_X is the lifetime of the excitation. The PL intensity is proportional to N_X/τ_X, i.e., it does also depend on the number of excitons N_X. Due to relaxation and scattering processes, which can take place prior to the recombination process, the excitons at lower energies may have a larger population than the excitons at higher energies. Therefore, in PL, the factor N_X may overcome a small oscillator strength $1/\tau_X$ and lead to a significant PL signal. Moreover, localization processes which may take place at low temperatures, can

lift the optical selection rules and thus cause radiative recombination processes. The effect of localization on dark states was recently discussed theoretically by Dzyubenko and Sivachenko.[26] A direct determination of the oscillator strength, however, can only come from an absorption measurement. Therefore, we have prepared optically thin samples. We were able to integrate external gates to the thinned samples, which allow us to tune the carrier density down to the dilute regime. This enabled us to measure the *direct* absorption of charged excitons at millikelvin temperatures in a dilution cryostat and compare it to PL. We were thus able to determine the oscillator strengths of the observed excitations. We believe that in our investigation we can clearly identify this charged exciton as a dark exciton of the GaAs quantum well for two main reasons. (i) In the optically thin samples no GaAs bulk material is left so that we can definitely rule out any bulk-related effects. (ii) By comparing PL *and* transmission at very low temperatures, we find that the oscillator strength of the excitation labeled X_{td}^- is close to zero which unambiguously shows that this is a dark mode.

The samples are one-sided modulation-doped GaAs–AlGaAs single quantum wells, consisting of a 25-nm-wide GaAs quantum well with a 88-nm AlGaAs spacer between the quantum well and the doped barrier region on one side, and a 100-period AlGaAs/GaAs (10 nm/3 nm per period) superlattice on the other side. On top of the sample, a 10-nm-thick titanium gate was deposited. The sample was glued upside down on a glass substrate using an UV curing optical adhesive. Subsequently, the sample was thinned from the back side by a selective etching process[27] down to the superlattice to a total thickness of about 1.3 μm. By applying a negative gate voltage, we can tune the density in the range between about 1×10^{10} cm^{-2} and 1×10^{11} cm^{-2}. PL and absorption measurements were performed via glass fibers in a ^{3}He/^{4}He dilution cryostat at a base temperature of $T = 40$ mK and magnetic fields up to 16 T. A sensor at the sample position indicated that during illumination the temperature directly at the sample is about $T = 100$ mK, while the base temperature is still $T = 40$ mK. Circularly polarized light was created directly inside the mixing chamber and left- and right-circularly polarized spectra were measured by ramping the magnet from positive to negative magnetic fields. For the PL, the sample was excited by a Ti:Sapphire laser at 750 nm. For the absorption measurement, a white light source was used.

Figure 1 shows a comparison of left-circularly polarized PL spectra, which were recorded at $T = 2$ K (upper spectrum) in an optical split-coil magnet, and at $T = 100$ mK (lower spectrum) in the dilution cryostat. In this polarization configuration, at $T = 2$ K, the well-known lower Zeeman components of the singlet exciton X_s^- and the neutral exciton X^0, and the bright triplet X_{tb}^- with parallel spin orientation of the electrons are visible. In the spectrum at $T = 100$ mK a strong line (labeled X_{td}^-) appears in between the X_s^- and the X_{tb}^-. We will argue below that this exciton is indeed a *dark* triplet exciton. Due to small sample inhomogeneities, the peaks in different measurement series do not always occur at exactly the same absolute positions. Therefore, in Fig. 1, the
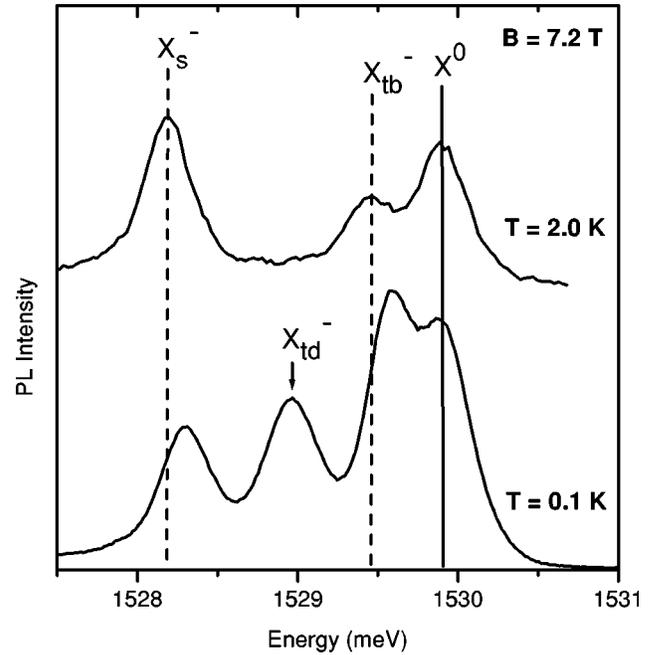

FIG. 1. Comparison of left-circularly polarized PL spectra for $T = 2$ K and $T = 0.1$ K. The electron density is in the range $n \approx (2 \pm 1) \times 10^{10}$ cm^{-2}.

spectrum at $T = 0.1$ K has been shifted by 0.16 meV so that the positions of the X^0 is the same in both spectra. For both spectra, the density is in the range $(2 \pm 1) \times 10^{10}$ cm^{-2}. We have recorded extensive series of spectra for different gate voltages in dependence on magnetic field. Throughout the whole magnetic field range, the X_{td}^- is not detectable at all at $T = 2$ K. For $T = 100$ mK, our finding is that the X_{td}^- is present in the spectra for filling factors $\nu < 1/3$, which is consistent with the results reported in Ref. 24. As examples, in Fig. 2, series of left-circularly polarized spectra are displayed for two different gate voltages, i.e., two different densities in the range of 10^{10} cm^{-2}. For the higher electron density [Fig. 2(a)] one can see that the neutral exciton X^0 is not visible in the displayed magnetic field range and the X_{td}^- splits from the bright triplet X_{tb}^- for $\nu < 1/3$. At significantly lower density [Fig. 2(b)], this splitting occurs at lower magnetic field [≈ 2 T in Fig. 2(b)], and, at high fields, the neutral exciton X^0 is visible. Consistent with the behavior predicted in Ref. 20, the binding energy of the X_{td}^- increases with magnetic field and the PL line approaches the X_s^- at high field [see Fig. 2(b)]. However, we note here that up to the highest field in our experiment (16 T) we did not observe the predicted singlet triplet crossing. We assume that it will occur at somewhat higher fields. According to the calculations in Ref. 22, the magnetic field value for the crossing should depend critically on the electron hole separation d perpendicular to the 2DES sheet. By comparing our experimentally observed binding energies of the X_{td}^- with calculations,[16,22] we can estimate for our structures $d < 0.5$ nm.

In the following we demonstrate that the observed X_{td}^- is indeed a *dark* mode. Figure 3 depicts absorption spectra in
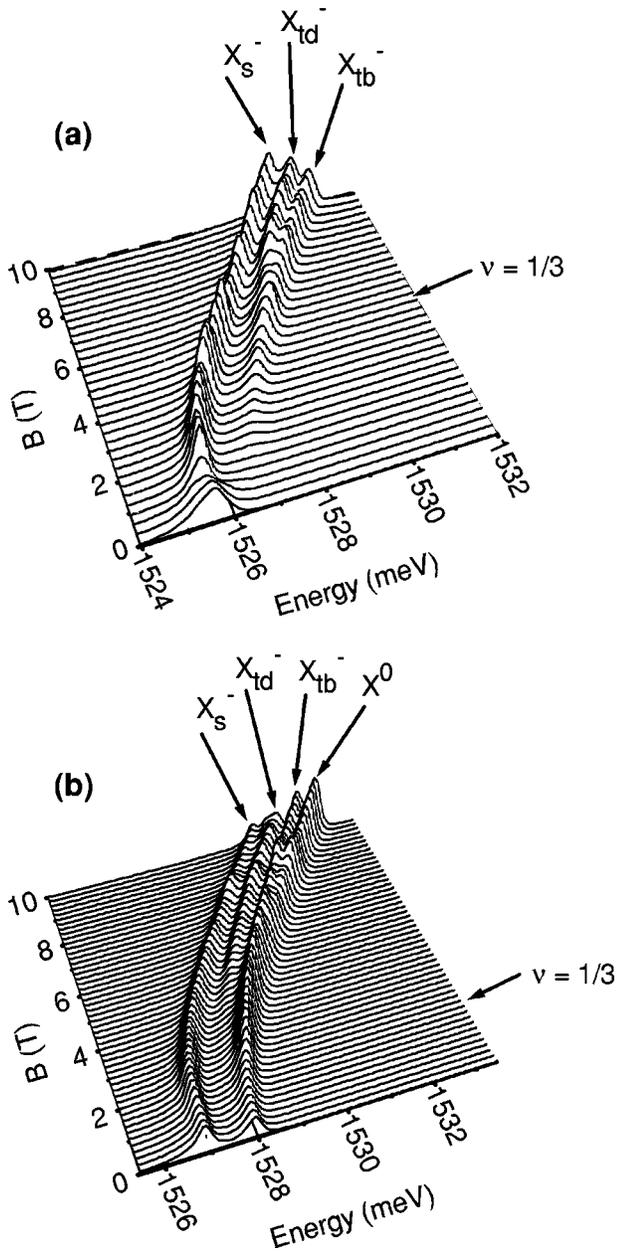
FIG. 2. Left-circularly polarized PL spectra for different magnetic fields for $T=0.1$ K and an electron density of (a) $n\approx4.8\times10^{10}$ cm^{-2}, and (b) $n\approx1.4\times10^{10}$ cm^{-2}.
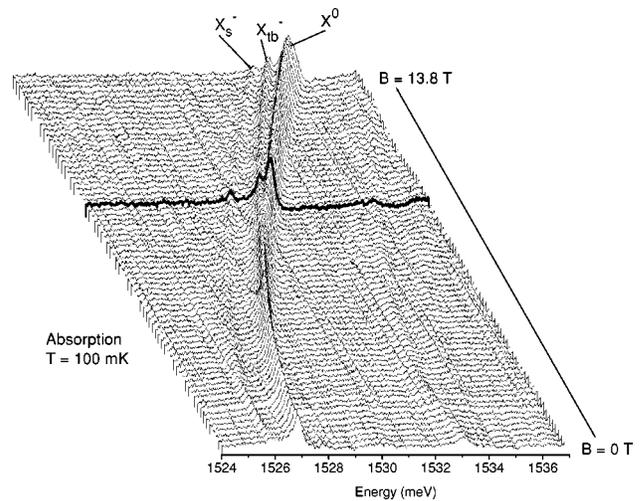

FIG. 3. Absorption spectra at $T=0.1$ K in dependence on magnetic field. For clarity, the spectra are shifted horizontally and vertically. The carrier density is about 2×10^{10} cm^{-2}. The inset shows a comparison of PL and absorption at $B=9$ T.

dependence on magnetic field at $T=100$ mK for an electron density of $n\approx2\times10^{10}$ cm^{-2}. One can clearly see absorption due to the *bright* excitons X_s^-, X_{tb}^-, and X^0. Remarkably, in Fig. 3 the neutral exciton X^0 has the highest absorption, i.e., the largest oscillator strength, which is proportional to $1/\tau_{X_0}$. As explained in the introduction, this is not directly reflected in the PL experiments (see Fig. 4), since the intensity of a PL line does via N_X/τ_X also depend on the number of excitons N_X, and on localization effects. From Fig. 4 one can see that the X_{td}^- does not appear in the absorption spectrum, though it exhibits a well developed line in the PL spectrum. From our signal-to-noise ratio we can estimate that the X_{td}^- has an oscillator strength which is at least one order of magnitude

smaller than the oscillator strength of X_s^-. This can be taken as a clear signature that the X_{td}^- is indeed a *dark* mode. According to Ref. 20, the total angular momentum of this mode is $\mathbf{L}=-1$. Therefore, in an idealized system, without scattering and localization, it cannot decay radiatively. This means, its lifetime τ should go to infinity and hence the oscillator strength $1/\tau$ to zero. This is directly reflected in the absorption spectra in Fig. 3. On the other hand, due to scattering or localization processes prior to recombination, in PL the selection rule $\mathbf{L}=0$ can be lifted. We believe that such scattering-assisted recombination can be the reason for the occurrence of X_{td}^- in the PL spectrum, though it is still puzzling why its PL strength is almost equal to those of the *bright* excitons. We think that our experimental observation, i.e., that the high PL intensity of the dark exciton is not of
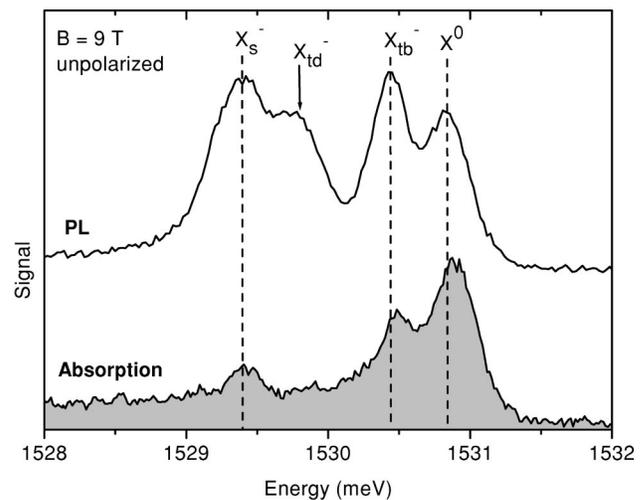

FIG. 4. Comparison of unpolarized PL and direct absorption spectra at $B=9$ T and $T=0.1$ K. The small cusp at $E=1529.9$ meV in the absorption spectrum is due to noise, resulting from the normalization procedure. This is also evident from Fig. 3.

intrinsic origin, is also very important for further theoretical treatments of this excitation. Furthermore, by our special sample preparation, which does not leave any GaAs bulk material in the sample, we can exclude impurity-related bulk excitons as a possible origin for the X_{td}^- line. (We note that for the spectra compared in Fig. 4, the PL was excited by a Ti:Sapphire laser and the absorption was measured using white light, which required different aligning procedures. Therefore, we cannot exclude slight shifts in the absolute positions of the observed lines due to sample inhomogeneities. This means that stokes shifts between absorption and PL cannot be seriously extracted from these measurements.)

In conclusion, by comparing PL and absorption spectra at very low temperatures, we could identify a dark triplet exciton X_{td}^- by showing that, in spite of a relatively high PL intensity, its oscillator strength is very small and justifies the assignment as a *dark* mode. We find that the X_{td}^- occurs at filling factors $\nu < 1/3$ only and is not visible at $T = 2$ K.

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